

DISCREPANCIES AND POSSIBLE ADJUSTMENTS IN THE 2200-m/s FISSION PARAMETERS

A. De Volpi



U of C-ALIA-USAEC

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) between the U. S. Atomic Energy Commission, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona	Kansas State University	The Ohio State University
Carnegie-Mellon University	The University of Kansas	Ohio University
Case Western Reserve University	Loyola University	The Pennsylvania State University
The University of Chicago	Marquette University	Purdue University
University of Cincinnati	Michigan State University	Saint Louis University
Illinois Institute of Technology	The University of Michigan	Southern Illinois University
University of Illinois	University of Minnesota	The University of Texas at Austin
Indiana University	University of Missouri	Washington University
Iowa State University	Northwestern University	Wayne State University
The University of Iowa	University of Notre Dame	The University of Wisconsin

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights.

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

DISCREPANCIES AND POSSIBLE ADJUSTMENTS IN
THE 2200-m/s FISSION PARAMETERS

by

A. De Volpi

Applied Physics Division

June 1971

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	7
I. INTRODUCTION.	7
A. Objectives	7
B. Circumstances	8
C. Controversial Data	9
II. STANDARD THERMAL CROSS SECTIONS (NONFISSION).	9
III. THERMAL-FISSION CONSTANTS	10
A. Surveys	10
1. Scattering Cross Sections.	11
2. Fission Cross Sections	11
3. Neutron Yield, ν	12
B. Values Recommended by IAEA.	12
C. Discrepancies in $\nu(^{252}\text{Cf})$	12
1. Review of $\nu(^{252}\text{Cf})$ Experiments	13
a. White and Axton	13
b. De Volpi and Porges	14
c. Axton	15
d. Comparison of Results	15
2. Least-squares Fit.	16
3. The Dilemma.	17
D. Weighting of Input Data.	18
E. Resolution of Dilemma	18
1. Manganese-bath Revisions for η	19
2. The Most Probable Value for $\nu(^{252}\text{Cf})$	20
a. Verified Values	20
b. Possible Systematic Discrepancy for Large Liquid-scintillator Measurements	22
c. Integral Experiments	23
3. $\sigma_f(^{235}\text{U})$	23
a. Effect of ^{233}U and ^{234}U Half-life Revisions	24
b. Measurements of $\sigma_f(^{235}\text{U})$	26
4. $\alpha(^{235}\text{U})$ and $\sigma_a(^{235}\text{U})$	28

TABLE OF CONTENTS

	<u>Page</u>
5. $\sigma_s(^{235}\text{U})$ and $\sigma_T(^{235}\text{U})$	29
6. Discordant Evidence	30
7. $\sigma_f(^{239}\text{Pu})$	30
F. Adjusted Values	32
1. ^{235}U	34
2. ^{233}U	35
3. ^{239}Pu	36
IV. DISCUSSION	37
A. Systematic Effects.	37
B. Role of Integral Measurements	38
C. Differential Experiments Needed	40
V. CONCLUSIONS.	41
REFERENCES.	43

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Values for 2200-m/s Constants Recommended by IAEA.	11
II.	Neutron Yield per Fission (ν_t) for ^{252}Cf	16
III.	Neutron Yield per Fission (ν) for Ratios to $\nu(^{252}\text{Cf})$	17
IV.	Neutron Yield per Fission (ν) for Ratios between ^{233}U , ^{235}U , and ^{239}Pu	17
V.	Neutron Yield per Absorption (η), Absolute Values	19
VI.	Half-life of ^{233}U	24
VII.	Half-life of ^{234}U	24
VIII.	Adopted Half-life Values	25
IX.	Fission Cross Sections	26
X.	Capture-to-Fission Cross-section Ratios (α).	28
XI.	Total and Absorption Cross Sections for ^{235}U	29
XII.	Scattering Cross Sections	29
XIII.	Fission Cross-section Ratios.	31
XIV.	Revised Values for 2200-m/s Constants	33
XV.	Estimated Accuracies of the Fission Parameters.	37

APPENDIX

Table

1	...
2	...
3	...
4	...
5	...
6	...
7	...
8	...
9	...
10	...
11	...
12	...
13	...
14	...
15	...
16	...
17	...
18	...
19	...
20	...
21	...
22	...
23	...
24	...
25	...
26	...
27	...
28	...
29	...
30	...
31	...
32	...
33	...
34	...
35	...
36	...
37	...
38	...
39	...
40	...
41	...
42	...
43	...
44	...
45	...
46	...
47	...
48	...
49	...
50	...
51	...
52	...
53	...
54	...
55	...
56	...
57	...
58	...
59	...
60	...
61	...
62	...
63	...
64	...
65	...
66	...
67	...
68	...
69	...
70	...
71	...
72	...
73	...
74	...
75	...
76	...
77	...
78	...
79	...
80	...
81	...
82	...
83	...
84	...
85	...
86	...
87	...
88	...
89	...
90	...
91	...
92	...
93	...
94	...
95	...
96	...
97	...
98	...
99	...
100	...

DISCREPANCIES AND POSSIBLE ADJUSTMENTS IN THE 2200-m/s FISSION PARAMETERS

by

A. De Volpi

ABSTRACT

The status of 2200-m/s fission constants has been reviewed, focused mainly on ^{235}U , ^{239}Pu , and ^{233}U . The most recent work in this field has been by the IAEA published in 1969. Depending very much on a partially subjective view toward experimental credibility, a significantly different set of parameters can be developed. The points of departure are in acceptance by the reviewer of the more recent low measurements of the ^{233}U and ^{234}U half-lives and of reduced down-weighting of some absolute measurements of $\nu(^{252}\text{Cf})$ which have been the object of extensive verification procedures. This results in 1% lower ν values for the fissile isotopes. In addition, some reductions in η values are experimentally justified. The half-life revisions augment evidence that the fission cross sections for ^{233}U and ^{235}U should be higher than the IAEA average. As a result of this study, an adjusted set of fission parameters is generated with substantive support from the revised experimental input data. A uniform constraint of constant product $\nu\sigma_f = \eta\sigma_a$ is applied, which makes most integral experiments insensitive to the modifications. Both ^{233}U and ^{239}Pu sets have additional ambiguities due to inadequate input data, indicating possible reductions in the constraining product.

I. INTRODUCTION

A. Objectives

To one not closely connected with the measurement processes, the wealth of nuclear data generated to improve reactor design could appear to be in a state of disarray. There are numerous conflicting results, many of recent vintage. This report provides an overview of the data that constitute the more fundamental fission parameters, the quantities basic to nuclear-reactor development. Included is a survey of various data compilations, conference proceedings, evaluations, and reviews. In the process of examining the available information, it is possible to construct a convergent pattern which indicates reasonable resolution of the apparent discrepancies.

Although thermal parameters have been relegated to a secondary role during the present period of fast-breeder development, these quantities provide the foundation upon which the remaining nuclear data are based. Thermal data retain an instrumental role in two ways. First, many cross sections at higher energies are measured relative to the reference point at 2200 m/s. Second, the quality of the thermal data reflects upon capabilities achievable at higher energies: Methods and the devices can often be tested in the better-established thermal environment. For the fundamental fission parameters, there is essentially no hope of achieving any accuracy greater than that acquired in the thermal standards.

In 1967, I reviewed the status of the relevant thermal parameters for Reactor and Fuel-processing Technology.²⁶ That paper is used as a building block for the present survey. The information will be updated with recent data and evaluations.

The chief fission parameters are the cross sections for fission (σ_f), capture (σ_γ), and scattering (σ_s) and the capture-to-fission ratio (α) along with the neutron yields per fission (ν) and per neutron absorbed (η). The dominant materials considered are ²³⁵U, ²³⁹Pu, and ²³³U. Although the various structural and coolant materials have a significant effect on breeding and operational characteristics, it is outside the scope of this report to discuss such an equally extensive subject area.

During the course of reviewing the current values available, it will be evident that discrepancies exist with varying significance. A further objective of the present overview is to provide recommendations for present use of the nuclear data which appear in reasonable agreement and to suggest alternatives where the data are discordant.

B. Circumstances

The 1967 review²⁶ reflected data available to a large extent through the first IAEA survey on 2200-m/s neutron constants (Westcott *et al.*⁷¹), and the first conferences on Neutron Cross Sections and Technology (1965), Physics and Chemistry of Fission (1965), and Nuclear Data for Reactors (1967). Subsequently, the 2200-m/s constants have been revised by the IAEA (Hanna *et al.*⁴²), and follow-up conferences have been held on cross sections (Washington, 1968; Knoxville, 1971), fission (Vienna, 1969), and nuclear data (Helsinki, 1970).

In addition, a consultants' meeting to discuss ν was held at Studsvik, Sweden, just before the Helsinki conference. At Studsvik the reference values for ²⁵²Cf and the thermal yields were discussed, as well as the resonance and higher-energy structure discovered in the neutron yield ν .

C. Controversial Data

In comparing the 1967 situation regarding nuclear data with the present 1971 status, one can observe the convergence of many quantities that had been poorly known. There are also residual problems still not adequately solved. Most notable, though, is the emergence of significant discrepancies in areas that were previously thought to be in satisfactory condition. The uncovering of these discrepancies is a result of and a tribute to continued evaluation of experiments, the associated techniques, and the constant pressure of refined measurements done by independent investigators using diverse methods. For accurate results at any level, verification by independent means is essential to attain a given level of meaningful confidence.

Regarding the 2200-m/s neutron constants for the four fissile nuclides, the 1969 IAEA survey⁴² recommends values with standard errors of a few-tenths of a percent on the primary constants; yet there are divergent data indicating that some of the parameters may be inaccurate to the extent of over 1%. The probable existence of a systematic error in the analysis is investigated, and a possible resolution of the dilemma is devised in the following pages of this report.

The ^{235}U fission cross section, aside from its direct utility, is the major fission reference from which ^{239}Pu and ^{233}U are standardized. Yet discrepancies of over 1% exist in the experimental data. Many intricacies are associated with an effort directed toward understanding the problem. Cross comparisons through related ratio measurements and a study of the standard cross sections used as a basis for the flux determinations are essential ingredients in an analysis of the dispersion.

A number of ratios of cross sections are useful in reactor applications: for example, $\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$ and $\sigma_f(^{238}\text{U})/\sigma_f(^{235}\text{U})$ as reaction-rate indices. Being relative quantities, such ratios are less subject to systematic error and thus are generally considered well known.

II. STANDARD THERMAL CROSS SECTIONS (NONFISSION)

Most thermal-cross-section measurements--otherwise absolute--depend upon some standard nonfission cross section. In particular, flux measurements accompanying fission-cross-section determinations have generally been carried out in the past by means of certain well-known reactions such as $^{197}\text{Au}(n,\gamma)$, $^{10}\text{B}(n,\alpha)$, and $\text{H}(n,p)$. Some thermal-fission cross sections rely upon the thermal values of the standards; the higher-energy measurements utilize the differential values of standards, usually calibrated against a flux or thermal-cross-section reference point.

Thus, we have an edifice of fission cross sections built upon absolute differential shape standards, which themselves are founded upon thermal calibrations. The 2200-m/s values for nonfission standards are well stabilized, and there is little to report that differs from the 1967 survey. There have been, however, important contributions to the shape standards in recent years.

An extensive study of the thermal standards was conducted by the IAEA team in 1965,⁷¹ and these values were revised as necessary in 1969.⁴² No change was needed in the reference $\text{Au}(n,\gamma)$ (98.7 ± 0.2 b), as two recent measurements were consistent with the adopted value. Some lesser-used cross sections were also evaluated: $\text{Co}(n,\gamma)$ at 37.50 ± 0.13 b, and $\text{Na}(n,\gamma)$ at 534 ± 5 mb.

Gubernator and Moret⁴⁰ have evaluated the $^{10}\text{B}(n,\alpha)$ standard, finding a best value of 3835 ± 7 b. A recent reanalysis of Argonne pulsed-neutron data by Meadows⁵⁷ gives a consistent measurement of 3842 ± 18 b.

An excessive range of measured values exists for the hydrogen absorption cross section. Published values now range from 321 ± 2.5 to 334.2 ± 0.5 mb, according to the IAEA survey,⁴² which chooses an average of 331 ± 4 mb.

For $^6\text{Li}(n,\alpha)$, Meadows' reexamination⁵⁷ of his own data provides 936 ± 4 b, of higher precision than the evaluation of Goldman *et al.*,³⁸ which listed 950 ± 15 b. Also, Uttley and Diment⁶⁸ extrapolated their total cross-section fit to a 2200-m/s value of 940 ± 6 b.

Goldman *et al.*³⁸ recommend an average of 5327 ± 10 b for the 0.0253-eV cross section of $^3\text{He}(n,p)$. Judging from the best accuracy available from other standards, the error should probably be increased to ± 15 b.

III. THERMAL-FISSION CONSTANTS

Much of the high-accuracy differential cross-section structure is contingent upon thermal standards. Moreover, there is sufficient redundancy in data and range of technique available for thermal-fission parameters that true consistency tests of accuracy can be made through variation of methods. Thus careful evaluation of the status of the fundamental fission constants at 2200 m/s provides knowledge of the limiting factors associated with measurements at higher energies, alongside the inherent value in precise knowledge of reference cross sections.

A. Surveys

The two primary surveys in this area have been supported by the IAEA in 1965⁷¹ and 1969.⁴² Each of the surveys is thorough and voluminous;

one purpose of the present report is to condense the results into a more concise representation. Another purpose is to provide a critical examination of some of the results obtained.

The second IAEA analysis includes new experimental data and uses modified 16-parameter least-squares fitting procedures for better treatment of measurements in Maxwellian spectra. Although 10 parameters have been determined with high precision for each of the major fissile nuclei (^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu) (see Table I), some significant problems have been noted by the IAEA group:

1. Scattering Cross Sections. There is a paucity of experimental data on scattering in fissile materials. Since thermal-neutron scattering is sensitive to the physical form of the target, there is also a complicated sample-dependent variation among the available data. At best, errors of $\pm 10\%$ may be attributed to the uranium isotopes and $\pm 20\%$ to the plutonium isotopes examined.

2. Fission Cross Sections. Explicit fission cross sections have always been more difficult to measure than total cross sections or some fission-parameter ratios. Exaggerating this difficulty, though, is the specific problem of determining the mass of fissile material used in the measurement. Those measurements of $\sigma_f(^{235}\text{U})$, which were dependent upon isotopic ratios coupled with detection of alpha activities of the ^{234}U component, are subject to an additional error due to serious disagreements which now exist in the half-life of ^{234}U . In addition, the decay constants for ^{233}U and ^{241}Pu are unsettled.

TABLE I. Values for 2200-m/s Constants Recommended
(cross sections in barns, g-factors for 20.4°C) by IAEA⁴²

	^{233}U	^{235}U	^{239}Pu	^{241}Pu
σ_a	577.6 ± 1.8	678.5 ± 1.9	1012.9 ± 4.1	1375.4 ± 8.6
σ_f	530.6 ± 1.9	580.2 ± 1.8	741.6 ± 3.1	1007.3 ± 7.2
σ_γ	47.0 ± 0.9	98.3 ± 1.1	271.3 ± 2.6	368.1 ± 7.8
α	0.0885 ± 0.0018	0.1694 ± 0.0021	0.3659 ± 0.0039	0.3654 ± 0.0090
η	2.2844 ± 0.0063	2.0719 ± 0.0060	2.1085 ± 0.0066	2.149 ± 0.014
ν_t	2.4866 ± 0.0069	2.4229 ± 0.0066	2.8799 ± 0.0090	2.934 ± 0.012
g_a	0.9965 ± 0.0013	0.9787 ± 0.0010	1.0752 ± 0.0030	1.0376 ± 0.0014
g_f	0.9950 ± 0.0021	0.9766 ± 0.0016	1.0548 ± 0.0030	1.0486 ± 0.0053
g_η	0.9985 ± 0.0017	0.9979 ± 0.0018	0.9810 ± 0.0027	1.0106 ± 0.0051
σ_s^{bound}	10.7 ± 1.8	17.6 ± 1.5	8.5 ± 2.0	12.0 ± 2.6

$$\nu_t(^{252}\text{Cf}) = 3.765 \pm 0.012$$

3. Neutron Yield, ν . The most pervasive problem is the continuing inconsistency in measured values of the neutron yield in fission, ν . Good direct measurements of the thermal yield for the major fissile isotopes are nonexistent. There are, however, ratio measurements in which the thermal values have been compared with each other or with the yield from ^{252}Cf . Thus $\nu(^{252}\text{Cf})$ takes a unique position as a standard for determining the thermal values.

Because the neutrons from ^{252}Cf arise out of spontaneous fission, it is easier to measure absolute values of its yield in the absence of reactor or accelerator neutron backgrounds or fluctuations. Yet, due to the necessity of making difficult absolute measurements in either or both of the fast-neutron and the fission-fragment occurrence rates, this parameter has so far defied efforts to obtain universally consistent results. Consequently, because the fission parameters σ_f , η , α , and ν are redundant, the thermal values of ν were drastically downweighted to the point of exclusion from significant influence upon the final IAEA output.

In connection with the inconsistencies among experimental results for ν , there are noticeable deficiencies in fission-neutron spectra. Differences of 10-15% exist in determinations of the average energy of neutrons coming from fission of the fissile materials and from ^{252}Cf , which is once again a convenient standard. Although the measurements of ν are, for the most part, not too sensitive to these uncertainties, such differences in magnitude complicate analysis of reactor spectrum-averaged cross sections.

B. Values Recommended by IAEA

Table I reproduces the current values for the 2200-m/s fission constants recommended by IAEA.⁴² With the prominent exception of α (the thermal capture-to-fission ratio), most of the differences with the original survey^{26,71} are modest. A 2.4% reduction in the ^{235}U value of α is suppressed in terms of influence on the remaining parameters, because the change in $1 + \alpha$ is only 0.33%. The apparent consistency of the two intensive studies has been interpreted as support for the validity of the data output.

On the other hand, the inconsistency between the input values derived from measurements of $\nu(^{252}\text{Cf})$ with the output values deduced through the least-squares fit is also a prominent feature of both studies. Thus an alternate interpretation to draw is that the least-squares fit reveals a persistent systematic discrepancy. Since the ν discrepancy is about 1%, this alternate interpretation suggests that some of the fission parameters in Table I are uncertain to the extent of 1%, even though their listed standard errors are just a few tenths of a percent.

C. Discrepancies in $\nu(^{252}\text{Cf})$

Some complications induced by discrepancies in measurements of $\nu(^{252}\text{Cf})$ have already been mentioned. The compl

serious than noted because this spontaneous fission isotope is the calibration standard for all other values of the neutron yield in thermal fission, and the thermal values of ν are the main reference points for the neutron yields at higher neutron energies. Thus, any absolute error in the value of $\nu(^{252}\text{Cf})$ carries over directly as an equivalent error in every value of the neutron yield at any energy for all fissile parameters.

In addition, there are integral measurement techniques in reactor physics which depend upon accurate knowledge of this quantity. The reactivity-oscillation method of determination of an integral value of $\bar{\alpha}$ for a zero-power critical facility (see Bretscher and Redman⁸) magnifies six-fold any neutron-yield inaccuracy for ^{252}Cf oscillated as a reference fission source. The null-zone technique, another type of measurement with the same objective, depends on ν for ^{235}U and ^{239}Pu to the same extent (see Till et al.⁶⁷).

Consequently, a detailed examination of the discrepancies that exist in the the neutron yield in fission of ^{252}Cf is justified by the wide-ranging importance of this quantity.

Of specific interest in the following discussion is the observation that there are essentially two separable sets of results, differing by 2-3%. Some measurements, especially those of the large liquid-scintillator type, give values close to 3.8 neutrons/fission; the other values tend to cluster around 3.7 neutrons/fission. Since each of these has been reported with standard errors in the range of 1% or better, it is immediately evident that the data spread exceeds a normal distribution.

1. Review of $\nu(^{252}\text{Cf})$ Experiments

A comprehensive review of the experiments has previously been provided.²⁶ Only those experiments postdating that review will be discussed in detail.

a. White and Axton. The preliminary value by White and Axton⁷² has since been published as 3.796 ± 0.031 for the total yield. The neutron-counting phase of this experiment is dependent upon the manganese bath facility at the National Physical Laboratory, Teddington, U.K., and the fission-fragment detection phase was conducted at the Atomic Weapons Research Establishment, Aldermaston, U.K. A recent report by White,⁷³ who carried out the fission counting, indicates that there is a 2% discrepancy between the low-geometry counting done at Aldermaston and the fission-rate evaluations done at Teddington. No reason was given for the difference.

If the fission-fragment rate determined at Aldermaston is 2% too low, then the reported value should have been 3.72 neutrons/fission. As a result, this measurement is equivocal. On the surface, it fits within the "high" group of values; in view of subsequent work by the experimenters, the value may belong in the "low" grouping.

b. De Volpi and Porges. A new measurement by De Volpi and Porges^{28,29} represents the only fully reported experiment since 1967. They have completely redone the fission-fragment and neutron-rate calibrations of the two fission counters previously reported;²⁶ in addition, they added a third fission counter, which provided about 99% fission-fragment detection efficiency. Thus the new reports encompass and supersede their previous results.

After anisotropy in neutron emission from fission fragments is taken into account, the yields from the three fission counters are quite consistent. Results from the fission counter with the highest efficiency dominate the final average. The fission rate from this counter was verified through defined low-geometry fission-fragment counting.

The neutron-detection phase of the experiment was done with the manganese bath, as before. Extensive independent investigations were carried out to verify the accuracy of the neutron-detection system. Among these investigations were concentration dependence on density,²⁷ high-energy parasitic capture in oxygen and sulfur, effective manganese-to-hydrogen absorption ratio, neutron escape, source and cavity self-absorption, and absolute detection of ⁵⁶Mn activity.

A number of sources having a wide range of emission spectra--Ra-Be(γ ,n), ²⁵²Cf, Ra-Be(α ,n), and Am-Be(α ,n)--were studied to unravel the energy-dependent nature of the correction factors.²⁸ In addition, two of the sources systematically studied were the international Ra-Be(α ,n) neutron standard NRC 200-1 and a United States standard Ra-Be(γ ,n), NBS-II. Excellent agreement with other calibrations was obtained, although discrepancies in some correction factors were noted.

The results of a limited international comparison in ⁵⁶Mn activity are now available.³⁰ Participants were able to count samples from common stock with a root-mean-square error of 0.1% for all results combined. The National Physical Laboratory (NPL) provided the samples.

Two additional reports substantiating correction factors for neutron escape³¹ and source and cavity absorption³² are published or accepted for publication. The results for the spontaneous fission spectrum of ²⁵²Cf are again bracketed by both more and less energetic neutron sources.

As a result of these investigations, the Argonne neutron assay is independent of unverified correction factors. In addition, the calibrations are corroborated through the international-standards comparison. The neutron measurements for soft-spectrum sources appear to differ with NPL by no more than 0.5%. This tends to provide an upper limit for the net effect of possible systematic deficiencies. Fortunately the various controversial correction factors in neutron escape, parasitic capture in sulfur and oxygen, and source and cavity absorption reach an overall minimum with ²⁵²Cf sources.

The neutron yield thus reported by Argonne²⁹ -- 3.725 ± 0.015 neutrons/fission (total yield) -- has been developed from absolute and verified procedures for both the neutron and fission rates.

c. Axton. At NPL, Axton is independently carrying out three sets of experiments on $\nu(^{252}\text{Cf})$. His results have not been published to date, but his preliminary values for the first set were available for the new IAEA survey,⁴² and his fission-fragment detection technique has been reported by Axton et al.³

For an initial sample containing 30% ^{252}Cf , Axton found a total yield of 3.700 ± 0.020 neutrons/fission.⁴² He reports⁴ that for a sample containing about 70% ^{252}Cf , his result is about 3.73 neutrons/fission, although the sample was too small for an accurate measurement. A larger sample of californium is available, and a new experiment is in progress.

As previously mentioned, there are strong connections between the neutron-detection capabilities of the NPL and the ANL facilities. However, there are significant differences in the fission-fragment detection method. Argonne uses an absolute neutron-fission coincidence technique, while NPL applies a model-dependent extrapolation method of fragment-fragment coincidence.³ Although the NPL technique places the extrapolation of fission-fragment detection efficiency to unity on a sturdier basis than simple extrapolation of single-channel fission-fragment rates, it continues to suffer from inherent limitations. Such a technique fails to satisfy one of the fundamental conditions of absolute, source-independent, coincidence calibration: namely, one detector uniformly sensitive to radiation emanating from throughout the source. (Another condition, a second detector with efficiency approaching 100%, is adequately satisfied.) As a result of the limitation, the required extrapolation is directly equal to the efficiency deficit.

d. Comparison of Results. A common denominator in all three of these recent measurements is the manganese bath. Since particular efforts have been made to compare sources, the neutron-assay phase may be considered to represent the most verified, hence least uncertain, aspect of the neutron-yield measurements. Moreover, the same facility was used for the neutron measurements of White and Axton⁷² and Axton (see Hanna et al.⁴²) just discussed. Since the preliminary neutron-yield values of Axton support the 1970 measurements of De Volpi and Porges,²⁹ their two independent fission-rate determinations appear to be in satisfactory agreement. This, then, is further reason to reevaluate the inconsistent fission-rate measurements reported by White and Axton;⁷² meantime, their reported value of 3.796 neutrons/fission must be downweighted.

Table II summarizes the more precise results obtained for $\nu_t(^{252}\text{Cf})$, as tabulated and adjusted by Hanna et al.⁴² The final ANL value

has replaced the preliminary result. Aside from the questionable datum of White and Axton,⁷² measurements based primarily on the manganese bath or having secondary calibrations by this method (e.g., Moat *et al.*⁵⁹ and Colvin *et al.*¹⁴) cluster together between 3.70 and 3.73. In addition, the entirely independent measurement of Colvin and Sowerby¹³ is within that low range. On the other hand, the two liquid-scintillator results are markedly high.

TABLE II. Neutron Yield per Fission (ν_f) for ^{252}Cf

Authors	Lab	Year	Reassessed Value ^a	Adopted Mean ^a	Reevaluated Value
Liquid Scintillator					
Asplund-Nilsson <i>et al.</i> ⁴²	FOA	1963	3.830 ± 0.037	3.807 ± 0.024	3.787 ± 0.037
Hopkins and Diven ⁴⁴	LAS	1963	3.793 ± 0.031		3.793 ± 0.031
Boron Pile Calibrated with d(γ,n)p					
Colvin and Sowerby ¹³	HAR	1965	3.713 ± 0.015	3.713 ± 0.024	3.713 ± 0.015
Dependent on NPL Manganese Bath					
Moat <i>et al.</i> ⁵⁹	ALD	1961	3.727 ± 0.056	3.713 ± 0.024	3.727 ± 0.056
Colvin <i>et al.</i> ¹⁴	HAR	1966	3.700 ± 0.031		3.700 ± 0.031
White and Axton ⁷²	ALD	1968	3.796 ± 0.031		3.796 ± 0.031
Axton <i>et al.</i> ³	NPL	1969	3.700 ± 0.020		3.700 ± 0.030
ANL Manganese Bath					
DeVolpi and Porges ²⁸	ANL	1969	3.725 ± 0.017	3.725 ± 0.024	3.725 ± 0.015
				Weighted mean:	3.740 ± 0.016
				Fitted value:	3.765 ± 0.010
Reevaluated weighted mean: ^b					3.731 ± 0.008

^aIAEA assessment.

^bWeighted mean without clustering.

The IAEA weighted average of all data is 3.740 ± 0.016 (revised by Hanna⁴³). The fitted value was that which Hanna *et al.*⁴² derived from the least-squares procedure, as compiled in Table I.

2. Least-squares Fit

The IAEA team⁴² subjected all available data on the major fissionable isotopes to a least-squares fit (LSF), the outcome of which is shown in Table II. In combination with the other data, the LSF generates a value of 3.765 ± 0.010 for the total yield of ^{252}Cf . More important to observe is that if all neutron-yield values were omitted from the input--which effectively removes the redundancy in the set of parameters--then the fissile isotopes combine to suggest the value $\nu_f(^{252}\text{Cf}) = 3.784 \pm 0.014$. This is the value for the neutron yield of ^{252}Cf deduced strictly from the cross sections, from η , from α values of the fissile nuclides, and from the ratios of ν for the fissile nuclides to that of ^{252}Cf --by omitting all absolute measurements of ν for ^{252}Cf .

Tables III and IV list the ν ratios used by Hanna *et al.*⁴²

TABLE III. Neutron Yield per Fission (ν) for Ratios to $\nu(^{252}\text{Cf})$
Delayed neutrons included. The second error is due to the uncertainties of fission-neutron spectra.

Authors	Lab	Year	$^{233}\text{U}/^{252}\text{Cf}$	$^{235}\text{U}/^{252}\text{Cf}$	$^{239}\text{Pu}/^{252}\text{Cf}$	Implied ^a $\nu(^{239}\text{Pu})$
Kenward <i>et al.</i> ⁴²	HAR	1958		$0.6458 \pm 0.0064 \pm 0$		
Meadows and Whalen ⁴²	ANL	1961		$0.6445 \pm 0.0114 \pm 0.0072$		
Hopkins and Diven ⁴⁴	LAS	1963	$0.6543 \pm 0.0079 \pm 0.0012$	$0.6437 \pm 0.0064 \pm 0.0012$	$0.7496 \pm 0.0090 \pm 0.0014$	2.797
Mather <i>et al.</i> ⁴²	ALD	1964	$0.6679 \pm 0.0082 \pm 0.0025$	$0.6380 \pm 0.0032 \pm 0.0024$	$0.7739 \pm 0.0090 \pm 0.0029$	2.887
Colvin and Sowerby ¹³	HAR	1965		$0.6423 \pm 0.0029 \pm 0$		
Condé ⁴²	FOA	1965		$0.6425 \pm 0.0056 \pm 0.0026$		
Fultz <i>et al.</i> ⁴²	LRL	1966	$0.6722 \pm 0.0106 \pm 0$	$0.6456 \pm 0.0212 \pm 0$		
Boldeman and Dalton ⁴²	AUA	1966		$0.6407 \pm 0.0013 \pm 0.0024$		
DeVolpi and Porges ²⁸	ANL	1969		$0.6445 \pm 0.0100 \pm 0$		
Weighted mean:			0.6635 ± 0.0052	0.6417 ± 0.0018	0.7618 ± 0.0067	2.842
Fitted values:			0.6604 ± 0.0018	0.6435 ± 0.0015	0.7648 ± 0.0022	2.853

^aBased on $\nu(^{252}\text{Cf}) = 3.731 \pm 0.008$.

TABLE IV. Neutron Yield per Fission (ν) for Ratios between ^{233}U , ^{235}U , and ^{239}Pu
Delayed neutrons included.

Authors	Lab	Year	$^{233}\text{U}/^{235}\text{U}$	$^{239}\text{Pu}/^{233}\text{U}$	$^{239}\text{Pu}/^{235}\text{U}$	Implied ^a $\nu(^{239}\text{Pu})$
Sanders ⁴²	HAR	1955	1.006 ± 0.020		1.179 ± 0.040	2.830
De Saussure and Silver ⁴²	ORL	1958	1.004 ± 0.012		1.173 ± 0.029	2.816
Colvin and Sowerby ¹³	HAR	1965	1.020 ± 0.006		1.182 ± 0.008	2.838
Boldeman and Dalton ⁷	AUA	1966	1.0281 ± 0.0033	1.1633 ± 0.0039	1.1960 ± 0.0044	2.871
Weighted mean:			1.0246 ± 0.0028	1.1633 ± 0.0039	1.1922 ± 0.0038	2.862
Fitted value:			1.0263 ± 0.0019	1.1582 ± 0.0025	1.1886 ± 0.0025	2.854

^aBased on $\nu(^{252}\text{Cf}) = 3.731$ and $\nu(^{235}\text{U}) = 2.400$.

3. The Dilemma

A marked dilemma is posed by the $\nu_t(^{252}\text{Cf})$ differences existing among:

- The LSF outcome of 3.784 ± 0.014 , independent of absolute measurements.
- The outcome of 3.765 ± 0.010 with no measurements excluded.
- The liquid-scintillator input values around 3.8.
- The manganese-bath and boron-pile input values near 3.7.

That the dilemma is becoming more formidable is justifiably concluded by noting that the most recent measurements of $\nu(^{252}\text{Cf})$ are close to 3.7 neutrons/fission. Yet the benefit of alertness to the difficulties of such absolute measurements was available, and each experiment represented the outcome of measurements upon several different samples of californium. On the other hand, data from ^{235}U and the other fissile isotopes, also measured absolutely, imply that $\nu(^{252}\text{Cf})$ should be about 2-3% higher.

Section E below offers a hypothesis that reconciles these discrepancies.

D. Weighting of Input Data

Some understanding of the weighting procedure in the least-squares-fit of Hanna et al.⁴² is useful for further discussion.

"The underlying basis of the least-squares procedure used in the present study is that the errors involved in the various measurements are of a random nature and are not correlated or systematic. It is also of fundamental importance to try to assess all errors on a uniform basis... The 'weight' of any value in the least-squares-fit is proportional to the inverse weight of its error, so that careful review is considered essential."

These were the general criteria used in the first study sponsored by the IAEA.⁷¹

As a result, each experimental input was examined in detail for its adequacy and uniformity of correction and for its completeness in terms of reporting. From this information, it was sometimes necessary to adjust results to a common basis, to apply retrospective corrections, or to down-weight because of inadequate documentation or because of other judgments.

The same general criteria were applied in the second IAEA study⁴² with further attention to uncertainties in neutron spectra.

In practice, for the least-squares fit, the dominant weights are given to absorption cross sections, to η and α values, to fission cross sections, and finally to ν values. It is the η values that essentially determine the neutron-yield outcome because of their relative significance in the weighting procedure. This importance arises because such experiments in measuring neutrons per absorption are considered relative neutron measurements (a ratio of fission neutrons issued to thermal neutrons absorbed), and rather small errors are assigned by the experimenters--around 1%. In contrast, the ν experiments require absolute measurements, although their reported errors have also typically been about 1%.

Thus the neutron yields recommended by the IAEA are dominated by the η measurements through the combined effects of relatively high weights to η and α input and reduced weight to ν .

E. Resolution of Dilemma

The statement regarding the underlying basis of the least-squares procedure quoted in Section D above acknowledges that the errors involved

in the various measurements must not be of a correlated or systematic nature. It is my hypothesis that there is indeed a systematic error pattern in the least-squares fit. To reconcile the dilemma posed by the diverse measurements, I propose that:

- The low values (~ 3.72) for $\nu_t(^{252}\text{Cf})$ are valid.
- The η average should be lowered by $3/4\%$.
- The ν values for the fissile isotopes should be about 1% lower.
- The fission cross sections should be over 1% higher.

The remaining discussion in this section develops the evidence for this reconciliation, which essentially preserves the product $\nu\sigma_f$ for ^{235}U as a constant. This would tend to explain why such substantial discrepancies in ν and σ_f could go undetected in view of the large amount of criticality data available from reactors fueled with ^{235}U .

1. Manganese-bath Revisions for η

There are just three absolute measurements of $\eta(^{235}\text{U})$. These are listed in Table V. The experiments by Macklin *et al.*⁵² and by Smith *et al.*⁶³ are based on the manganese bath for neutron detection. That of Gwin and Magnuson⁴¹ is independent of the bath.

TABLE V. Neutron Yield per Absorption (η), Absolute Values

Authors	Lab	Year	Original Input			Suggested Revisions		
			^{233}U	^{235}U	^{239}Pu	^{233}U	^{235}U	^{239}Pu
Macklin <i>et al.</i> ⁵²	ORL	1960	2.288 ± 0.013	2.076 ± 0.013		2.268	2.058^c	
Macklin <i>et al.</i> ⁴²	ORL	1962			2.118 ± 0.019			2.100
Gwin and Magnuson ⁴¹	ORL	1962	2.2735 ± 0.0185^a	2.076 ± 0.0211^a		2.2735	2.076	
Smith <i>et al.</i> ⁶³	MTR	1966	2.298 ± 0.0122	2.079 ± 0.0157	2.108 ± 0.0104	2.289	2.071^d	2.100
		Weighted mean:	2.2896 ± 0.0080^b	2.0770 ± 0.0091^b	2.1103 ± 0.0091	2.278	2.067	2.100
		Fitted value:	2.2844 ± 0.0056	2.0720 ± 0.0053	2.1085 ± 0.00585			
Vidal <i>et al.</i> ⁶⁹	CEN	1970	2.240 ± 0.012	2.072 ± 0.006				
Magnuson ⁵³	ORL	1970	2.283 ± 0.015					2.106
			2.292					

^ag-dependent data, errors excluding g-errors.

^bMean value composed of g-dependent data.

Effect upon Reported Value of η

	Resonance Absorption	Neutron Escape	Parasitic Capture
c	-0.7%	+0.3%	-0.5%
d	-0.2%	+0.3%	-0.5%

Recent measurements with manganese baths at Argonne and NPL¹ indicate that three of the correction factors applied for the two manganese-bath measurements should be revised. The corrections are for neutron escape from the tank, for parasitic capture in sulfur and oxygen, and for resonance absorption in manganese. The first two

corrections are each of the order of a few-tenths of a percent, and they tend to cancel each other. Thus the dominating effect is that of resonance manganese capture.

Direct measurements of leakage have been made³⁰ using ^{252}Cf as a source, whereas the original corrections were based on Monte Carlo calculations not substantiated by direct experiment. The sulfur/oxygen effect has been derived by De Volpi and Porges,²⁸ also by direct experiment with ^{252}Cf . There are experiments and calculations that disagree and others that agree with these numbers; fortunately, the effects approximately cancel each other, as mentioned.

The resonance absorption correction is by Axton and Ryves,² and it differs only slightly from an earlier work by Axton, Cross, and Robertson¹ and somewhat more from data by De Juren and Chin.²⁰

To be consistent with existing practice, it is necessary to append these three corrections of the manganese bath, resulting in the numbers listed for ^{235}U in Table V under "Suggested Revisions." Comparable percentage changes should be made to the other fissile isotopes.

The resulting revised weighted mean is 1% less than the previous experimental average, 1/4% below the fitted value, whereas previously it was 1/4% above the fitted value.

2. The Most Probable Value for $\nu(^{252}\text{Cf})$

Given the diversity of numbers reported by experimenters, as discussed in Section III.C and displayed in Table II, can a best value for $\nu(^{252}\text{Cf})$ be selected on the basis of a critical examination? The answer can be "yes," if the following factors are given sufficient weight:

- a. That the low values are valid, since those experiments with the greatest degree of verification agree in this regard.
 - b. That there is a systematic discrepancy in the large liquid-scintillator measurements.
 - c. That the experiment of White and Axton⁷² contains an error in fission assay.
 - d. That various integral measurements tend to support the lower values.
- a. Verified Values. In Table II, those measurements subjected to extensive verification procedures have not been accorded proportionately high weights. Unable to resolve the conflicting data, the IAEA team⁴² allotted equal weights to four categories of measurements, as reflected under "Adopted Mean." Yet several of these measurements have undergone substantial supplementary effort not credited in the downweighting process.

The liquid-scintillator measurements have not been the subject of any follow-up studies reported in the literature, although Condé^{17,18} has continued to investigate some systematic effects.

The boron pile, on the other hand, was examined in detail for systematic response (see Colvin *et al.*¹⁴) up to the time it was dismantled. The results of a series of calibrations using standard neutron sources are listed in Table II under "Dependent on NPL Manganese Bath."

The neutron-detection phase of the measurements by Axton has been the beneficiary of numerous cross-checks with worldwide neutron standards. The $\nu(^{252}\text{Cf})$ measurement done by White and Axton⁷² should be downweighted because of internal discrepancies in fission-fragment detection, noted in Section III.C. In addition, the ν values derived strictly at NPL have not been fully documented and require a fission-detection extrapolation procedure so far unverified in published work. As a result, the adopted weight of 0.75% may be reasonable.

The measurement of De Volpi and Porges, as described in Section III.C, has been carried out with absolute measurements, by independent methods, each verified for both the fission²⁹ and neutron²⁸ determinations. Special supplementary investigations were made specifically for ^{252}Cf fission-neutron effects. Moreover, extensive corroboration was obtained through comparisons with international neutron source standards.

Except for the two most recent measurements, all the $\nu(^{252}\text{Cf})$ determinations involve single samples. De Volpi and Porges made use of three samples of differing origin during a period of two years, each sample being calibrated under somewhat varied conditions. Axton also has had recourse to three samples, the results of the first one being released in the preliminary account.⁴

Having a multiplicity of californium samples provides a basis for tests on internal consistency and on systematic sample-related effects. This is particularly helpful for the fission-fragment calibration phase.

Comparing the "Reassessed Value" error with the "Adopted Mean" error in Table II shows that the IAEA-attributed error does not reflect the magnitude of verification effort associated with the experiments mentioned above. The errors associated with the liquid-scintillator results are each about 1%; yet the combined error is taken to be 0.65%.

The boron-pile measurements were downweighted from 0.4 to 0.65%. The manganese-bath measurements of De Volpi and Porges, reported by the authors at 0.4%, were also downweighted to 0.65%.

The net effect of these error reassessments has been to artificially produce a weighted mean of 3.740 ± 0.016 . A weighted mean of all "reassessed values" (restoring the error of 0.015 for De Volpi and Porges) yields $\nu(^{252}\text{Cf}) = 3.731 \pm 0.008$, even without further deemphasis of the White and Axton measurement.

This relatively low value arises only on the basis of the experimenter's assessed error. It does not take into further account either the relative degree of verification, the sample multiplicity, nor the fact that the three most recent measurements have a weighted mean of 3.715 ± 0.009 .

In view of the relatively high degree of consistency among $\nu_t(^{235}\text{U})/\nu_t(^{252}\text{Cf})$ ratios (see Table III), the anticipated value of $\nu_t(^{235}\text{U})$ would be $0.6417 \times 3.731 = 2.394$ neutrons/fission. The only direct experiment for $\nu_t(^{235}\text{U})$ not otherwise included as a ratio is a measurement by Kenward *et al.*⁴⁸ reassessed by Hanna *et al.*⁴² to be 2.398 ± 0.033 .

b. Possible Systematic Discrepancy for Large Liquid-scintillator Measurements. In view of the tendency for the most recent and the most verified values of $\nu(^{252}\text{Cf})$ to be about 2-3% less than those derived through the use of the large liquid-scintillator systems, it is logical that more attention is now being paid to a search for errors in this area.

Two effects have recently been studied: delayed gamma rays, and a gamma-multiplicity-related neutron-efficiency dependence. Both of these, if not fully taken into account in the original work, would reduce the reported value by a retrospective correction.

Studies by Walton and Sund⁷⁰ give a measure of isomeric transitions that could lead to gamma rays during the gate times used for neutron slowing-down. For ^{239}Pu and ^{235}U , the possible errors in ν_p range from 0.8 and 0.6% down to zero, depending on the bias level chosen for the gamma-ray detection. Although these numbers imply that ν_p ratio measurements against a ^{252}Cf standard are likely to be unaffected, it is possible that for the absolute measurement of ^{252}Cf , such an effect could produce a uniform overcount in the neutron channel. Data for delayed gammas from ^{252}Cf are presently insufficient, although all calculations suggest a negligible influence.

The most recently uncovered possibility for bias in liquid-scintillator results has arisen from some methodical studies by Soleilhac and colleagues.⁶⁴ Their examination of the behavior of system response as a function of gamma-ray detection efficiency indicates that as much as a 1.5% correction should be made in neutron efficiency to take into account gamma-ray multiplicity effects in their large liquid scintillator. It is not possible to apply these tests to the other liquid-scintillator systems under

original conditions; however, Condé¹⁸ can reproduce an effect of magnitude 0.6%, while Mather⁵⁵ sees a much smaller influence. Since Hopkins and Diven⁴⁴ use a system with high gamma and neutron efficiency (89%), their results are less likely to be sensitive to such a discrepancy. On the other hand, the effect may be geometry and bias dependent, which necessitates specific investigation in each system under the reported measurement conditions.

Until there is further evaluation of both of these effects, there is little hope of resolving the difficulties presently thrust upon the liquid-scintillation data. In any event, these are two plausible sources of systematic deviation, which combined could be responsible for two inaccurate values of $\nu(^{252}\text{Cf})$.

c. Integral Experiments. A third aspect that tends to support relatively low values of ^{252}Cf is some indirect results from integral experiments in lower-power reactors. One current technique for measurements of integral values for the capture-to-fission ratio $\bar{\alpha}$ averaged over a reactor core is to oscillate a sample of ^{252}Cf as a representative insertion of fission neutrons. The number of neutrons per fission is a parameter of the experiment. Experiments on French reactors (Barré⁶⁵) tend to produce better agreement with differential data when a low ^{252}Cf value is applied in the calculations. A similar conclusion may be drawn from U.S. experiments (Bretscher et al.⁹). Both of these experiments magnify the effect of the neutron-yield dependence, so that the difference between the high and low values (3.7 to 3.8) would be reflected by 15% effects on $\bar{\alpha}$.

There are also reported to be some integral experiments (Casini¹¹) which concur with lower values for the neutron yield in the fissile isotopes. Since the ratios of the neutron yields for fissile isotopes versus ^{252}Cf are not, for the most part, subject to such a large uncertainty, the lower values in the fissile isotopes imply a lower value for ^{252}Cf and conversely.

3. $\sigma_f(^{235}\text{U})$

If the neutron yield for ^{252}Cf is as low as the preceding analysis indicates, then $\nu(^{235}\text{U})$ would have to be reduced significantly below the value of 2.423 given in Table II. This is independent of the 1/2% reduction in $\eta(^{235}\text{U})$ previously considered. Without some readjustment of the remaining fissile parameters, an incompatibility with reactor criticality experience would arise. There is, however, an escape from the criticality constraint: It is primarily the product $\nu\sigma_f = \eta\sigma_a$ that must be conserved. Thus a reduction in ν must be tested against an equivalent increase in σ_f .

In this section we evaluate fission cross-section data, particularly to see if there is fully independent evidence to support a corresponding increase in the accepted σ_f average.

Before making an explicit study of the cross-section measurements, let us inspect the status of ^{233}U and ^{234}U half-lives.

a. Effect of ^{233}U and ^{234}U Half-life Revisions. The isotopes ^{233}U and ^{234}U play an important role in certain aspects of fission cross-section measurements. For highly enriched ^{235}U it is sometimes useful to spike a solution with ^{233}U for the purposes of quantitative analysis (Keith *et al.*⁴⁷); otherwise, the ^{234}U content can be used as an indication of total uranium content when the isotopic analysis is well known (Deruytter and Becker²³). Tables VI and VII contain a tabulation of measured half-lives

TABLE VI. Half-life of ^{233}U

Half-life, 10^5 yr	Author (Year); Comments
$1.63 \pm ?$	Linenberger ⁴² (1945)
1.62 ± 0.01	Hyde ⁴² (1946)
1.615 ± 0.004	Sellers <i>et al.</i> ⁴² (1953)
1.603 ± 0.008	Bigham <i>et al.</i> ⁶ (1958)
1.626 ± 0.008	Dokuchaev and Osipov ⁴² (1959)
1.615 ± 0.009	Popplewell ⁴² (1961); see Keith ⁴⁶ below
1.6210 ± 0.0032	Ihle <i>et al.</i> ⁴⁵ (1967)
1.540 ± 0.003	Oetting ⁶⁰ (1967)
1.554	Same, when corrected for ^{232}U daughters
1.553 ± 0.010	Keith ⁴⁶ (1968); Popplewell ⁴² and this work used the same calibrated low-geometry counters.
1.588 ± 0.007	Durham ⁴² (1969); preliminary value

TABLE VII. Half-life of ^{234}U

Half-life, 10^5 yr	Author (year); Comments
2.69 ± 0.27	Nier ⁴² (1939)
2.29 ± 0.14	Chamberlain <i>et al.</i> ⁴² (1946)
2.35 ± 0.14	
2.69 ± 0.04	Goldin <i>et al.</i> ⁴² (1949)
2.520 ± 0.008^a	Kienberger ⁵⁰ (1949/1952)
2.475 ± 0.016	Fleming <i>et al.</i> ⁴² (1952)
2.519 ± 0.025	Bigham <i>et al.</i> ⁶ (1958); calculated from the data on "enriched uranium"
2.47 ± 0.03	White <i>et al.</i> ⁴² (1965)
2.444 ± 0.005	De Bievre <i>et al.</i> ¹⁹ (1970)
2.439 ± 0.014	Meadows ⁵⁶ (1970)

^aError should be increased to at least 0.033 on basis of experimenter's judgment of accuracy of measurement.

compiled by Hanna *et al.*,⁴² with some preliminary values confirmed and a new measurement by Meadows⁵⁶ added. As indicated by the braces, there have been major decreases in the measured values of the ²³³U and ²³⁴U half-lives within recent years. Table VIII summarizes the half-lives adopted by Hanna *et al.*,⁴² plus two selected values of highest precision.

TABLE VIII. Adopted Half-life Values

Nuclide	Half-life, yr	Percent Accuracy	Selected Values
²³³ U	1.593×10^5	± 1.5	$1.554 \pm 0.003 \times 10^{5a}$
²³⁴ U	2.488×10^5	± 0.64	$2.444 \pm 0.005 \times 10^{5b}$
²³⁵ U	7.10×10^8	± 1.5	
²³⁹ Pu	2.438×10^4	± 0.2	
²⁴¹ Pu	14.5	± 3	
Natural uranium	$\frac{\text{Specific activity}}{1504.6 \text{ dpm/mg}}$	± 0.5	

^aWeighted mean of Oetting⁶⁰ (1967) and Keith⁴⁶ (1968) measurements in Table VI.

^bWeighted mean of three most recent published measurements in Table VII.

Selection of the half-lives for ²³³U and ²³⁴U, rather than straightforward development of the weighted mean, is a departure from normal practice which requires justification. A weighted mean is valid when there is no improper error assignment or when there is no suspicion of systematic error. For ²³³U, the case for a possible systematic error is barely defensible in view of the liquid-scintillation determination of specific activity by Ihle *et al.*⁴⁵ However, the usually reliable calorimetric method of Oetting⁶⁰ suggests a fundamental discrepancy. Of the three most recent entries, Keith⁴⁶ has confirmed the lower value of Oetting,⁶⁰ and Durham's measurement was given by Hanna *et al.*⁴² as a private communication without documentation. As a result, the "selected value" of $1.554 \pm 0.003 \times 10^5$ years for the half-life of ²³³U must be recorded as a premise to be tested by its rectifying action upon otherwise diverse data. The most arbitrary feature of the "selected value" is the error assignment, coming directly from Oetting's work.⁶⁰ The IAEA⁴² assigns a 1.5% error "compromise," which better reflects the uncertainty in the specific disintegration rate for ²³³U.

In the case of ²³⁴U (which here is of far greater importance than ²³³U), the selection task is founded on an improved basis. There is a large spread in the values before 1965. The three measurements since then are in rather good agreement. Of major significance is the high degree of technique variation, reproducibility (with specified limitations), and thoroughness that can be attached to the work at CBNM (De Bierre *et al.*¹⁹). Thus, the "selected value" is dominated by this well-qualified accurate measurement, although the selection arises from a weighted mean

of the three most recent values. One should observe that the one other measurement (Kienberger^{49,50}) with a small error (0.32%) is listed only with Kienberger precision. Kienberger also mentions an accuracy of at least 1%, which is consistent with the limitations of the 2- π counting technique.

Deruytter *et al.*²² reported a preliminary measurement of $\sigma_f(^{235}\text{U})$ at the 1968 Washington Cross Section and Technology conference. To illustrate the effect of the difference between the "adopted" and the "selected" values, the calculated change in this cross section is from 576.8 to 587.3 b, corresponding to ^{234}U half-lives of 2.488×10^5 and 2.444×10^5 years.

b. Measurements of $\sigma_f(^{235}\text{U})$. Table IX contains a full listing of the fission cross-section measurements of the three main fissile isotopes, taken from Hanna *et al.*⁴² with a column added containing "revised values" based on the "selected values" of Table VIII for the ^{233}U and ^{234}U decay constants.

TABLE IX. Fission Cross Sections (barns)

Authors	Lab	Year	^{233}U	^{235}U	^{239}Pu	^{235}U Revised Value	^{233}U Revised Value
Popovic and Grimeland ⁴²	KJL	1953		589.7 \pm 13.5 ^a		589.7	
Popovic and Saeland ⁴²	KJL	1955	536.3 \pm 18.8 ^a			585.8	536.3
Raffle ⁴²	HAR	1955	507.5 \pm 21.8	585.8 \pm 22.3	702.1 \pm 23.9	589 \pm 11	507.5
Friesen <i>et al.</i> ³⁶	HAN	1956		557.0 \pm 14			
Bigham <i>et al.</i> ⁶	CRC	1958	523.51 \pm 6.85 ^a		742.81 \pm 5.35 ^a		540.0
Saplaoglu ⁶²	ANL	1958		602.6 \pm 10.3		602.6	
Deruytter ²¹	MOL	1961		590.0 \pm 8		590.0	
Maslin <i>et al.</i> ⁵⁴	ALD	1965		572.0 \pm 7		572.0	
Keith <i>et al.</i> ⁴⁷	ALD	1968	538.67 \pm 6.33 ^a	583.77 \pm 11.14 ^a	741.92 \pm 6.75 ^a	583.8	538.7
Deruytter <i>et al.</i> ²²	GEL	1968		577.0 \pm 7.6		587.9 \pm 3.4	
Additional error for discrepant input data:				\pm 1.73			
Weighted mean:			530.95 \pm 4.42 ^b	581.58 \pm 3.87 ^b	741.26 \pm 4.13 ^b		
Fitted values:			530.6 \pm 1.65	580.2 \pm 1.6	741.6 \pm 2.8		
Frayse and Prosdocimi ³⁵	CEA	1965				596.5 \pm 10	
Deruytter and Becker ²³	GEL	1970			742.5 \pm 3.7		
Revised weighted mean:					742.5 \pm 2.8	587.4 \pm 2.5	539.3 \pm 4.6

^ag-dependent data, errors excluding g-errors.

^bMean value composed of g-dependent and independent data.

The measurement by Friesen *et al.*³⁶ is so sparsely documented that I have excluded it from the list of "revised values"; also it is nearly three standard deviations from the weighted mean.

The value of Deruytter and Becker²³ has a dominant weight because of the small error based on their own estimate. Nevertheless, the weighted mean of the "revised values" excluding their measurement is still 586.8 ± 3.6 b.

In the report by Hanna *et al.*,⁴² the error given by Deruytter and Becker²³ initially was expanded by $\sqrt{2}$ because the work had then been reported only in preliminary form. Inasmuch as final results were given at the Helsinki Conference on Nuclear Data, the original estimate by Deruytter and Becker can be retained. The work by Deruytter and Becker²³ is particularly noteworthy because of their extensive effort on sample preparation, analysis of sample weights, use of redundant targets, and application of low-geometry counting. Also of great importance is that the measurement was performed on a slow chopper through the energy range of 0.005 to 0.1 eV. Precision low-geometry alpha counting is considered a significant improvement over 2π and 4π counting. The measurement by Deruytter and Becker²³ was based on a ^{10}B normalization, whereas an earlier and consistent result of Deruytter²¹ referred to the gold cross-section standard. This earlier value is independent of alpha disintegration constants.

Also incorporated is the datum by Fraysse and Prosdocimi,³⁵ omitted by Hanna *et al.*⁴² because of a possible systematic error in the interpretation of the boron counting data. Adjustment for the ^{234}U half-life raises the reported value of Fraysse and Prosdocimi to 596.5 b.

The intermediate result by Keith *et al.*⁴⁷ is based on the lower ^{233}U half-life, directly measured by Keith.⁴⁶ Because the cross sections were determined in a thermal-reactor flux, they cannot be given as high a weight as the monochromatic measurements. The capture cross section of ^{59}Co was used for a standard.

Examination of the "revised values" demonstrates a high degree of uniformity in the range of 587 b.

Of the recent measurements, only that of Maslin *et al.*⁵⁴ continues to be low; it is also among those that are independent of alpha counting for sample assay. On the other hand, the technique applied is similar to that originated by Saplakoglu,⁶² which yields a significantly high value. Experience indicates (see De Volpi and Porges²⁵) that difficulty can occur in obtaining uniform beam conditions to justify some of the assumptions required for the method. As a result of the magnification in fission rate produced by the booster, it appears possible to underestimate the systematic errors involved in the technique.

In any event, the error assigned to the measurement by Maslin *et al.*⁵⁴ seems unduly small for a precision experiment since the rms deviation for the 45 and 90° data averaged together with equal weights is ± 23 b, in contrast to their weighted error of ± 6 b.

If one accepts the lower ^{234}U half-life, then the weighted mean for the 2200-m/s fission cross section of ^{235}U becomes 587.4 ± 2.5 b. Since the evidence for the reduced half-life is rather strong, any procedure

not taking this into consideration on a uniform basis can lead to a systematic bias. The $1\frac{1}{4}\%$ difference between this revised mean and the fitted values of Hanna et al.,⁴² as repeated in Table IX, may reflect such a deficiency.

4. $\alpha(^{235}\text{U})$ and $\sigma_a(^{235}\text{U})$

Although there are large disagreements outside of quoted errors among the measured capture-to-fission ratios (see Table X), the net influence within the least-squares fit is not too significant for ^{235}U (and ^{233}U) because of the relatively small value of α . The measurement of Lounsbury et al.,⁵¹ has a weight equal to the combined weight of all other ^{235}U experiments, and their α ratios for ^{233}U and ^{239}Pu strongly dominate the weighted mean. Of all the ^{235}U fissile parameters, the 2.4% reduction in weighted mean for α from the 1965 value⁷¹ is the largest revision.

TABLE X. Capture-to-Fission Cross-section Ratios (a)

Authors	Lab	Year	^{233}U	^{235}U	^{239}Pu
Inghram <u>et al.</u> ⁴²	ANL	1955	0.0943 ± 0.0029^a		
Cornish ⁴²	HAR	1960		0.187 ± 0.0140^a	
Okazaki <u>et al.</u> ⁴²	CRC	1964	0.0904 ± 0.0011^a	0.1687 ± 0.0015^a	
Cabelli ⁴²	HAR	1966	0.0867 ± 0.0020^a	0.1678 ± 0.0026^a	0.3504 ± 0.0140^a
Durham <u>et al.</u> ³³	CRC	1967		0.1727 ± 0.0015^a	0.3572 ± 0.0059^a
Cabelli ⁴²	HAR	1968			0.3735 ± 0.0180^a
Lisman and Rider ⁴²	MTR	1968	0.0939 ± 0.0020^a	0.1693 ± 0.0015^a	
Conway ⁴²	BET	1968	0.0845 ± 0.0042^a	0.1660 ± 0.0072^a	
Lounsbury <u>et al.</u> ⁴²	CRC	1969	0.0899 ± 0.0004^a	0.1702 ± 0.0007^a	0.3601 ± 0.0021^a
Weighted mean:			0.09001 ± 0.00036^a	0.17011 ± 0.00053^a	0.35975 ± 0.00195^a
Fitted values:			0.0885 ± 0.0016	0.1694 ± 0.0019	0.3659 ± 0.0035
Weighted mean excluding Durham <u>et al.</u> ³³ (1967) and Lounsbury <u>et al.</u> ⁴² (1969)				0.1688 ± 0.0010	
Average of all except Cornish ⁴² (1960) measurement; with rms error				0.1691 ± 0.0021	

^ag-dependent data, errors excluding g-errors.

As a result of deemphasis in the direct ν input data, the output value of ν in the IAEA fitting process is determined chiefly from the relationship $\nu = \eta(1 + \alpha)$. Then a 2.4% reduction in α is diminished by the term $\alpha/(1 + \alpha)$ to have an effect of 0.35% on ν . On the other hand, there are other input data for the absorption cross section, so that the α input has a small influence on σ_a but a major role in estimating σ_c because of the lack of direct capture cross-section experiments.

A value of $\alpha(^{235}\text{U}) = 0.17011 \pm 0.00053$ implies, by itself, $\sigma_a(^{235}\text{U}) = 687.3 \pm 2.7$ b, based on the higher component for fission determined in Section 3 above. This compares with the experimental weighted mean of 679.46 ± 2.54 b found by Hanna et al.,⁴² which closely agrees with the values used in 1965.^{26,71} There have been no new measurements of the absorption cross section since the 1965 study.

Using the fitted value of Hanna *et al.*⁴² for $\alpha(^{235}\text{U}) = 0.1694$, one obtains $\sigma_a(^{235}\text{U}) = 686.9 \pm 2.7$ b.

5. $\sigma_s(^{235}\text{U})$ and $\sigma_T(^{235}\text{U})$

One of the most reliable measurements that can be made in nuclear physics is that of the total cross section.²⁶ From such determinations, the absorption cross section is obtained by subtracting the scattered fraction. Herein lies another difficulty. Having deduced a total cross section that is 7.8 b higher than previously accepted and being confined by intrinsically reliable measurements of the total cross section, I reexamined the reported values of the scattering cross section.

To Table XI, taken from Hanna *et al.*,⁴² I have added a column indicating the values of scattering cross section subtracted to produce the absorption result. Table XII lists the measured scattering cross sections given by Hanna *et al.*

TABLE XI. Total and Absorption Cross Sections for ^{235}U (barns)

Authors	Lab	Year	Authors' σ_T	Reassessed σ_T	Sample	σ_a	σ_s Subtracted
Egelstaff ³⁴	HAR	1957	724 \pm 15	724 \pm 26	Metal	708.25 \pm 26.4	15.7
Melkonian <i>et al.</i> ⁴²	COL	1953	691 \pm 5	694 \pm 14	Rolled metal	678.7 \pm 14.6	15.3
Palevsky <i>et al.</i> ⁴²	BNL	1954	700 \pm 5	700 \pm 10	Metal	684.25 \pm 11.1	15.7
Nikitin <i>et al.</i> ⁴²	ITE	1955	710 \pm 20	Accepted	Unknown, not liquid	692.0 \pm 21.2	18.0
Simpson <i>et al.</i> ⁴²	MTR	1959	690 \pm 10	690.0 \pm 9.6	Metal	674.25 \pm 12.2	15.7
Safford <i>et al.</i> ⁶¹	COL	1959	694.97 \pm 1.81 698.68 \pm 4.81	696.0 \pm 2.5 698.68 \pm 5.1	Liquid Rolled metal	679.0 \pm 2.5 683.38 \pm 6.65	17.0 15.3
Block <i>et al.</i> ⁴²	ORL	1960	693 \pm 5	Accepted	Rolled metal	677.7 \pm 6.58	15.3
Saplaoglu ⁴²	ANL	1961	694.2 \pm 1.5	696.0 \pm 2.5	Rolled metal	680.7 \pm 4.95	15.3
Gerasimov and Zenkevich ³⁷	KUR	1962	670 \pm 8 (σ_a)	670.0 \pm 10.0 (σ_a)		670.0 \pm 10.0	
Common partial error for bound-atom scattering:						± 1.7	
Weighted mean:						679.46 \pm 2.54	
Fitted value:						678.5 \pm 1.7	

TABLE XII. Scattering Cross Sections (barns)

	^{233}U	^{235}U	^{239}Pu	^{241}Pu
Potential Scattering:	12.0 \pm 2.0	11.5 \pm 1.1	10.2 \pm 1.1	12.0 \pm 2.2
Sample				
Liquid (bound atom)	11.6 \pm 2.3	17.0 \pm 1.7	8.6 \pm 2.1	12.0 \pm 2.6
Powdered or sintered metal	11.6 \pm 3.4 (± 2.51)	17.0 \pm 3.8 (± 3.40)	8.6 \pm 2.7 (± 1.70)	12.0 \pm 3.5 (± 2.35)
Rolled metal	10.5 \pm 3.8 (± 3.03)	15.3 \pm 4.6 (± 4.28)	7.7 \pm 3.0 (± 2.14)	10.8 \pm 4.0 (± 3.04)
Metal, not specified	10.85 \pm 4.15 (± 3.45)	15.75 \pm 5.05 (± 4.76)	8.0 \pm 3.3 (± 2.55)	
Oxide	14.6 \pm 5.5 (± 5.0)	20.0 \pm 5.3 (± 5.0)	11.6 \pm 5.4 (± 5.0)	15.0 \pm 5.6 (± 5.0)
Unknown, not liquid	13.4 \pm 6.7 (± 6.29)	18.0 \pm 7.3 (± 7.1)	10.85 \pm 6.15 (± 5.78)	
Rolled metal ^a		14.3 \pm 0.5		

Note: Values in parentheses are partial errors due to the crystal structure.
^aCeulemans and Poortmans.¹²

Ceulemans and Poortmans¹² have made the first measurement of the (rolled-metal) scattering cross section at 0.0253 eV, whereas previous data tabulated in Table XI represented, at best, extrapolations from 0.27 eV. The new measurement of 14.3 ± 0.5 b is 1 b less than that used for rolled-metal foils in Table XI. Accordingly, an absorption cross section about 1 b greater than the IAEA value would appear to be reasonable, since metal foils predominate in the transmission measurements.

In looking over the list of absorption cross-section results, one finds that the liquid-sample work of Safford *et al.*⁶¹ is strongly weighted in comparison with all other measurements. A simple unweighted average of the absorption values is 681.3 b, omitting the outlying values of Egelstaff³⁴ and Gerasimov and Zenkevich³⁷ and treating the liquid and rolled-metal data of Safford *et al.* as independent measurements. Uniformly replacing a 1-b scattering cross section would bring this average up to 682.3 b.

6. Discordant Evidence

As indicated in examining total cross-section limitations, the evidence does not conclusively support a $1\frac{1}{4}\%$ increased fission cross section. It becomes necessary to invoke additional hypotheses so that some combination of the following could apply:

- a. $\alpha(^{235}\text{U})$ should be lower than the present indicated average. This could account for as much as 2 or 3 b in σ_f .
- b. The scattering cross section is even smaller than measured by Ceulemans and Poortmans.¹² Perhaps 1 b could be obtained from this stipulation.
- c. Errors in the measured transmission cross sections and the deduced absorption cross sections allow about 2.5 b for a single standard deviation.

Thus there are no exclusive arguments to account for an additional 6-7 b needed to sustain a consistent theory; instead, from the observed data, one must resort to statistical inference which permits a reasonably high probability of overlap with the higher fission cross section previously derived. The error estimates, to any reliable degree, do not exclude the theory that revision of the ^{235}U fission cross section is warranted.

7. $\sigma_f(^{239}\text{Pu})$

Potentially, a further difficulty exists in terms of some of the $^{239}\text{Pu}/^{235}\text{U}$ fission ratios. The numbers compiled by Hanna *et al.*⁴² are reproduced in Table XIII, supplemented with the recent datum of Deruytter and Becker.²³ There appears to be strong evidence that the weighted mean

and fitted values are too high. The high weighted mean arises from the strong influence of the measurement of Lounsbury *et al.*,⁵¹ which, because of its reliance upon mass-spectrometric methods and independence of alpha half-lives, would appear to be justified.

TABLE XIII. Fission Cross-section Ratios

Authors	Lab	Year	$^{233}\text{U}/^{235}\text{U}$	$^{239}\text{Pu}/^{233}\text{U}$	$^{239}\text{Pu}/^{235}\text{U}$
Raffle ⁴²	HAR	1955	0.8664 ± 0.0597	1.3834 ± 0.0853	1.1986 ± 0.0656
Bigham <i>et al.</i> ⁶	CRC	1958	0.9141 ± 0.0012^a	1.4189 ± 0.0088^a	
Frayse and Prosdoci ³⁵	SAC	1965			1.268 ± 0.023^a
White <i>et al.</i> ⁴² (Monokinetic)	ALD	1966			1.253 ± 0.030
(Thermal)					1.252 ± 0.030^a
Keith <i>et al.</i> ⁴⁷	ALD	1968	0.9227 ± 0.0069^a	1.3773 ± 0.0229^a	1.2709 ± 0.0343^a
Lounsbury <i>et al.</i> ⁵¹	CRC	1969	0.9208 ± 0.0068^a	1.4060 ± 0.0092^a	1.2947 ± 0.0104^a
		Weighted mean:	0.9145 ± 0.0012^b	1.4101 ± 0.0061^b	1.2818 ± 0.0083^b
		Fitted values:	0.9145 ± 0.0024	1.3976 ± 0.0056	1.2781 ± 0.0049
Bigham <i>et al.</i> ^{6c}					1.256 ± 0.013
Deruytter and Becker ⁴²	GEL	1970			1.2629 ± 0.0099
Vidal <i>et al.</i> ⁶⁹	CEN	1970	0.914 ± 0.005		
Weighted mean including Lounsbury <i>et al.</i> ⁵¹					1.2740 ± 0.0064
Weighted mean excluding Lounsbury <i>et al.</i> ⁵¹					1.2615 ± 0.0081

^ag-dependent data, errors excluding g-errors.

^bMean value composed of g-dependent and independent data.

^cRevised to ^{233}U half-life of 1.553×10^5 years.

There is also a measurement by Bigham *et al.*,⁶ excluded from the IAEA review because of systematic redundancy with the other ratios. Lounsbury *et al.*⁵¹ quote the value at 1.2970 ± 0.0075 to demonstrate that their relatively high result is consistent with a previous Canadian irradiation by some of the same authors. However, as noted by Hanna *et al.*,⁴² the work of Bigham *et al.* is directly proportional to the $^{233}\text{U}/^{239}\text{Pu}$ half-life ratios. If one adopts the ^{233}U half-life measured by Keith,⁴⁷ then the Bigham *et al.* $^{233}\text{U}/^{239}\text{Pu}$ and $^{235}\text{U}/^{239}\text{Pu}$ ratios need to be reduced by about $1.553/1.620$, giving the revised value added to Table XIII. As a result, the latest NRU measurement stands alone; a weighted average including it gives the ratio at 1.2740 ± 0.0064 ; omitted, the ratio becomes 1.2615 ± 0.0081 , which differs from the Lounsbury *et al.* datum by over four standard deviations.

Referring back to Table IX, the fission cross sections found for ^{239}Pu cluster about the weighted mean of 741.3 ± 4.1 b, especially when the recent value of Deruytter and Becker²³ is introduced. This, in fact, changes the weighted mean to 742.5 ± 2.8 b. All three of these measurements are independent of the ^{233}U and ^{234}U disintegration-constant difficulties.

The table of fission ratios (Table XIII) includes some additional values for the $^{239}\text{Pu}/^{233}\text{U}$ quotient. With the original weighted mean of 1.2818 combined with the 741.3-b cross section for ^{239}Pu of Table IX, a ^{235}U fission cross section of 578.3 b is obtained, strongly influencing the least-squares

multiparameter fit of Hanna *et al.*⁴² This would have been considered discordant evidence with regard to the hypothesis of a higher ^{235}U fission cross section. Now, however, in light of the reassessment provided above, the weighted mean for $^{239}\text{Pu}/^{235}\text{U}$ fission ratio (which excludes the recent Canadian experiment), combined with the updated ^{239}Pu fission cross section, warrants a value of $\sigma_f(^{235}\text{U}) = 588.6 \pm 4.1$ b. This now becomes supportive evidence for a higher evaluation of the ^{235}U fission probability.

F. Adjusted Values

An outgrowth of the preceding analysis, an independent assessment of the fundamental fission parameters can be constructed. The adjusted values recommended for consideration are collected in Table XIV. The fission parameters for the three major isotopes are included. Plutonium-241 has been omitted, but those modifications that occur across the board are relevant to that isotope as well.

The columns labeled "Experiment" are either the IAEA experimental average used as input to their least-squares fit⁴² or revised values arising from this review. The columns labeled "Adjustment A," "Adjustment B," "Adjustment C," and "Adjusted" are the result of a juggling process in which the experimental parameters were confined by the restraints $\nu\sigma_f = \eta\sigma_a = \eta(1+\alpha)\sigma_f = c$. In particular, the constant c was identical to that developed by Hanna *et al.*,⁴² except for "Adjustment B" in the ^{233}U and ^{239}Pu sets. These cases will be discussed separately below.

In parentheses, under the experimental and adjusted values, are percentage deviations from the appropriate experimental average or least-squares output of the IAEA (see Table I). Those labeled 0% are taken directly from the work of Hanna *et al.*⁴² Positive percentages are for values larger than the reference work.

The errors associated with the input experimental quantities are usually derived from errors in the weighted means, in a manner similar to Ref. 42. Errors appended to the adjusted values are also from Hanna *et al.*, for the most part. Since the least-squares fitting procedure does not recognize systematic error possibilities, the output errors are essentially unaffected by adjustments that reflect systematic influences. These errors are "optimistic" in that the estimates do not consider the fact that the changes recommended greatly exceed the previously assigned errors. Thus the original error values did not provide a correct guide to the accuracy of the fission parameters. On the other hand, one of the main virtues in the revised values of Table XIV, with regard to the ^{252}Cf standard, is a higher degree of internal consistency, especially with ^{235}U , thereby tending to justify the coupled standard deviations.

A fundamental point of departure for the revisions can be traced to two partially subjective decisions: I have selected values of the half-lives for ^{233}U and ^{234}U as indicated from the latest experimental data; Hanna *et al.*⁴²

TABLE XIV. Revised Values for 2200-m/s Constants

233U				235U		239Pu			
Experiment	Adjustment A ^a	Adjustment B ^a	Adjustment C ^b	Experiment	Adjusted	Experiment	Adjustment A ^c	Adjustment B ^c	
σ_a	575.6 ± 1.6 (0%) ^d	582.5 ± 1.8 (+0.85%)	578.0 (+0.07%)	575.6 (-0.35%)	680.5 ± 2.7 (+0.15%)	683.0 ± 1.9 (+0.66%)	1012.1 ± 6.2 (0%)	1021.6 (+0.86%)	1013.4 ± 4.6 (+0.13%)
σ_f	539.3 ± 4.8 (+1.6%)	537.9 ± 1.9 (+1.4%)	536.5 (+1.1%)	531.9 (+0.25%)	587.4 ± 2.5 (+1.0%)	585.7 ± 1.8 (+0.95%)	742.5 ± 2.8 (+0.26%)	742.5 (+0.26%)	742.5 ± 3.1 (+0.26%)
σ_γ	50.6 ± 3.2 (0%)	44.6 ± 0.9 (-5%)	41.5 (-13%)	43.7 (-7.6%)	-	97.3 ± 1.1 (-1.0%)	275.5 ± 7.8 (0%)	279.1 (+2.9%)	270.9 ± 2.6 (-1.7%)
α	0.0900 ± 0.0004 (0%)	0.0830 ± 0.0018 (-6.6%)	0.0773 (-14%)	0.0822 (-7.7%)	0.1691 ± 0.0021 (-0.59%)	0.1661 ± 0.0021 (-2.0%)	0.3598 (0%)	0.376 (+2.8%)	0.365 ± 0.004 (+1.4%)
η	2.278 ± 0.008 (-0.44%)	2.265 ± 0.006 (-0.86%)	2.278 (-0.29%)	2.284 (0%)	2.067 ± 0.009 (-0.48%)	2.058 ± 0.006 (-0.68%)	2.100 ± 0.009 (-0.48%)	2.091 (-0.84%)	2.091 ± 0.007 (-0.84%)
ν_t	2.464 ± 0.005 (-1.0%)	2.453 ± 0.007 (-1.4%)	2.454 (-1.3%)	2.472 (-0.6%)	2.393 ± 0.008 (-1.2%)	2.400 ± 0.007 (-0.92%)	2.854 ± 0.008 (-1.0%)	2.877 (-0.10%)	2.854 ± 0.007 (-0.91%)
σ_s^e	10.5 (+24%)	3.5 (-59%)	8.1 (-4.9%)	10.5 (+24%)	14.3 ± 0.5 (-7.0%)	13.6 ± 1.5 (-5.0%)			
$\nu_t(^{252}\text{Cf}) = 3.731 \pm 0.008$ (Experiment) (-0.35%) = 3.735 ± 0.008 (Adjusted) (-0.94%)									
$\sigma_f(^{239}\text{Pu})/(^{235}\text{U}) = 1.2633 \pm 0.0081$ (Experiment) (-1.5%) = 1.2677 ± 0.0081 (Adjusted) (-0.8%)									
$T_{1/2}(^{233}\text{U}) = 1.554 \pm 0.003 \times 10^5$ years (Adjustment A) (-2.5%) = 1.562 ± 0.003 × 10 ⁵ years (Adjustment B) (-2.0%)									
$T_{1/2}(^{234}\text{U}) = 2.444 \pm 0.005 \times 10^5$ years (Experiment) (-1.8%)									
$\nu_t(^{235}\text{U})/(^{252}\text{Cf}) = 0.6414 \pm 0.0018$ (Experiment) (0%) = 0.6426 ± 0.0015 (Adjusted) (0%)									
$\nu_t(^{239}\text{Pu})/(^{252}\text{Cf}) = 0.7648 \pm 0.0067$ (Experiment) (0%) = 0.7703 ± 0.0022 (Adjustment A) (+0.85%)									
$\nu_t(^{235}\text{U})/\sigma_f(^{235}\text{U}) = 1405.6$ (Experiment) (-0.01%) = 1405.7 (Adjusted) (-0.01%)									
$\eta(^{235}\text{U})/\sigma_a(^{235}\text{U}) = 1406.6$ (Experiment) (+0.06%) = 1405.6 (Adjusted) (-0.01%)									
$\nu_t(^{233}\text{U})/\sigma_f(^{233}\text{U}) = 1328.8$ (Experiment) (+0.71%) = 1319.5 (Adjustment A) (0%) = 1316.6 (Adjustment B) (-0.22%)									
$\eta(^{233}\text{U})/\sigma_a(^{233}\text{U}) = 1311.2$ (Experiment) (-0.63%) = 1319.4 (Adjustment A) (0%) = 1316.7 (Adjustment B) (-0.21%)									
$\nu_t(^{239}\text{Pu})/\sigma_f(^{239}\text{Pu}) = 2119.1$ (Experiment) (-0.81%) = 2136.2 (Adjustment A) (+0.02%) = 2119.1 (Adjustment B) (-0.81%)									
$\eta(^{239}\text{Pu})/\sigma_a(^{239}\text{Pu}) = 2125.4$ (Experiment) (-0.51%) = 2136.2 (Adjustment A) (+0.02%) = 2119.0 (Adjustment B) (-0.81%)									
Other Values									
$\eta(^{239}\text{Pu})/\sigma_a(^{239}\text{Pu})/\eta(^{235}\text{U})/\sigma_a(^{235}\text{U}) = 1.5110$ (Experiment) (+0.15%) = 1.5197 (Adjustment A) (+0.03%) = 1.5075 (Adjustment B) (-0.76%)						1.509 ± 0.023 (Magnuson ⁵³)			
$(\eta - 1)/(^{239}\text{Pu})/\sigma_a(^{239}\text{Pu})/(\eta - 1)/(^{235}\text{U})/\sigma_a(^{235}\text{U}) = 1.5333$ (Experiment) (+3.6%) = 1.5424 (Adjustment A) (+0.09%) = 1.5300 (Adjustment B) (-0.90%)									
$\eta(^{239}\text{Pu})/\eta(^{235}\text{U}) = 1.0160$ (Experiment) (+0.13%) = 1.0160 (Adjustment A) (-0.16%) = 1.0160 (Adjustment B) (-0.16%)						1.017 (Vidal et al. ⁶⁹)			
$\eta(^{233}\text{U})/\eta(^{235}\text{U}) = 1.1021$ (Experiment) (-0.05%) = 1.1006 (Adjustment A) (-0.17%) = 1.1069 (Adjustment B) (+0.40%)						1.081 ± 0.005 (Vidal et al. ⁶⁹)			
$\eta(^{233}\text{U})/\sigma_a(^{233}\text{U})/\eta(^{239}\text{Pu})/\sigma_a(^{239}\text{Pu}) = 0.9322$ (Experiment) (-0.19%) = 0.9386 (Adjustment A) (0%) = 0.9367 (Adjustment B) (-0.20%)						0.934 ± 0.014 (Magnuson ⁵³)			

Other Values

1.509 ± 0.023 (Magnuson⁵³)1.017 (Vidal et al.⁶⁹)1.081 ± 0.005 (Vidal et al.⁶⁹)0.934 ± 0.014 (Magnuson⁵³)^aFor ²³³U Adjustment A, $\nu_t(^{233}\text{U})/\sigma_f(^{233}\text{U}) = 1319.5$ (as Hanna et al.⁴²); for Adjustment B, the product is 1316.6.^bFor $\nu_t(^{233}\text{U})/\sigma_f(^{233}\text{U}) = 1314.8$.^cFor ²³⁹Pu Adjustment A, $\nu_t(^{239}\text{Pu})/\sigma_f(^{239}\text{Pu}) = 2136.2$ (as Hanna et al.⁴²); for Adjustment B, the product is 2119.1.^dIn parentheses are percentage differences comparing Hanna et al.⁴² input-experimental and output-adjusted data. (The experiment averages derived in this report are compared with IAEA experimental averages; the adjusted output is compared with the IAEA LSF.)^eRollled metal.

chose to average all the experimental work together as shown in Table VIII. Second, the IAEA group give a very large weighting to the NRU/NRX α -irradiation measurements, whereas I have looked upon their work as subject to common characteristic deficiencies. In this regard, examine Table X and the accompanying discussion. It is immediately apparent that the measurements are very sensitive to the various g-factors. For example, Hanna *et al.*⁴² reduced the result of Durham *et al.*³³ by 2.2%. The datum of Cabell¹⁰ underwent a 0.6% increase. It thus becomes almost mandatory to conclude that complications in conversion of reactor temperature spectra to Maxwellian spectra and then to 2200 m/s are too uncertain to warrant more than a few percent confidence in irradiation measurements of α , an impression sustained by comparing the 1965 survey⁷¹ with the 1969 reevaluation.⁴² In particular, the recommended values of the IAEA for α were reduced by 2.4% in the transition from the 1965 to the 1969 surveys, while the experimental input average dwindled by 1.9%.

A second fundamental point of departure has been in assessment of the $\nu(^{252}\text{Cf})$ average. Although the mean value used (3.731 ± 0.008 neutrons/fission) is just slightly under the IAEA average, I have given this undiminished weight in the data set; Hanna *et al.*⁴² on the other hand, drastically downweighted this input datum.

As a result, it was necessary for Hanna *et al.* to deduce the ν values for the fissile isotopes from the remaining parameters, using the α measurements as a major factor. In this review, I have reversed the procedure, applying the redundant data to infer a possible inaccuracy in α .

Remarks specific to the three main fissile isotopes are covered in the following three subsections.

1. ^{235}U

The value of c , as defined earlier in this section, conserved for ^{235}U is shown in Table XIV to be 1405.7 (neutrons/fission)(barns/atom). There is no available external evidence to detract from this value. This constraint causes a slight reduction in the deduced fission cross section, although still nearly 1% above the IAEA results. Also, the η output undergoes an additional reduction over that suggested from the manganese-bath revisions. Since no direct input data were used in the ν_t values (the quality of direct ν experiments on the fissile isotopes is relatively poor), there is no comparison basis for the experimental input. However, the output value remains almost 1% below the IAEA recommendation. There are additional 1-b reductions in the capture and scattering cross sections caused by the constraints. The largest adjustment is in the capture-to-fission ratio, which requires a 2% reduction in order to rectify the overdetermined data set. As discussed in Section E.4 above, this is not inconsistent with previous trends in the same direction.

The "adjusted" value for $\nu(^{252}\text{Cf})$ reflects the "adjusted" values for the three fissile isotopes, as well as the experimental averages for the relative yields with respect to ^{252}Cf . Since the errors in the neutron-yield ratios are comparable to the error in the absolute californium yield, this feedback is marginally justified. A least-squares refitting would probably split the differences more uniformly, as well as taking into account the effects from the various yield ratios with better precision.

It was necessary to increase the absorption cross section by an additional $1/2\%$ in order to develop an internally consistent set. This adjustment, reasonably within error limits of the experiments and within possible values of the scattering cross section, does not appear to be excessive. If the scattering cross section were uniformly reduced by 1 b, which can be carried out as an extension of the lower rolled-metal cross section found by Ceulemans and Poortmans,¹² and if all absorption cross sections were given equal weight, then the adjusted value of $\sigma_a(^{235}\text{U})$ is consistent with the experimental average as revised.

It is pertinent to observe that a $1/2\%$ increase in the absorption cross section causes the Gwin and Magnuson⁴¹ value of $\eta(^{235}\text{U})$ to be decreased by about $1/4\%$, a move consistent with the additional reductions in η during the transition from the experimental average to the adjusted values.

2. ^{233}U

The combination factor c for ^{233}U was initially held at the IAEA value of 1319.4. Increases in the absorption and fission cross sections appear justified, while the neutron yields per fission and per absorption go through rather large decreases in comparison with IAEA indications. However, significantly less capture offsets the reactivity losses caused by fewer neutrons emitted.

Adjustment A, based on the constraint $c(^{233}\text{U}) = 1319.5$, implies a rolled-metal scattering cross section of 3.5 b compared to the Hanna et al.⁴² input value of 10.5 ± 4.8 b and output value of about 8.5 ± 1.8 b. To avoid being more than one standard deviation from these more plausible IAEA numbers, one possible combination of adjustments, labeled B, is based on the following operations: (1) Reduce the constraining constant $c(^{233}\text{U})$ by 0.2%; (2) increase $\eta(^{233}\text{U})$ at the expense of lowered $\sigma_a(^{233}\text{U})$ and $\alpha(^{233}\text{U})$; and (3) decrease $\sigma_f(^{233}\text{U})$ slightly more. The resulting scattering cross section is 8.1 b. The partially arbitrary nature of these manipulations reflects an approximate degree of uncertainty associated with the ^{233}U parameters.

Incidentally, the reduced value of the ^{233}U fission cross section suggests that the ^{233}U half-life may be about $1/2\%$ larger than the "selected value" from Table VIII; such an increase is more compatible with a weighted mean of all experimental values.

Recent studies at Westinghouse provide compelling evidence of shape factors that strongly influence the experimental input data for ^{233}U . The changes affect fission, capture, and neutron production. If the spectrum-associated adjustments are applied according to the calculations of Steen⁶⁶ and Mitchell and Emert,⁵⁸ then the ^{233}U parameters would have (1) slightly lower experimental input averages for the absorption and fission cross sections, (2) over 1% lower α , and (3) 0.4% higher η . The Steen analysis is based on a value of the constraint $\eta\sigma_a$ reduced 0.76% to 1309.3. With a less drastic reduction in the constraint ($c = 1314.8$), Adjustment C in Table XIV is calculated.

3. ^{239}Pu

There is ambiguity in the possible output values for ^{239}Pu . Since there has been little basis for modifications to the experimental averages--other than the common reductions in ν and η --there has been no compensation by possible increases in the fission cross sections. Thus, to retain the value of c used by Hanna *et al.*,⁴² the "Adjustment A" column applies. A primary feature of this combination is to increase ν to a level similar to that suggested by the IAEA and also to increase the capture and absorption probabilities.

The available experimental evidence discourages an increase in the absorption cross section, but otherwise does not resist the other effects. A specific examination of the neutron-yield ratios (see Table IV) involving ^{239}Pu is also ambiguous. The most recent measurements by Boldeman and Dalton⁷ on the $^{239}\text{Pu}/^{235}\text{U}$ ratio are consistent with $\nu_t(^{239}\text{Pu}) = 2.870$. Also, their ratio for $^{233}\text{U}/^{235}\text{U}$ gives a value of $\nu_t(^{233}\text{U})$ that is in agreement with the adjusted values of Table XIV. On the other hand, the Boldeman and Dalton data are relatively low for the ratio $\nu_t(^{235}\text{U})/\nu_t(^{252}\text{Cf})$ (see Table III).

On the basis of the ^{235}U data previously derived, the weighted mean from Table IV suggests $\nu_t(^{239}\text{Pu}) = 2.861$. This is chiefly an outcome of the high weight attached to the Australian measurements.⁷

One further point of ambiguity arises from the $\nu_t(^{239}\text{Pu}/^{252}\text{Cf})$ data of Table III. There are two values in wide variance with each other. The lower one gives $\nu_t(^{239}\text{Pu}) = 2.796$, which is substantially below the values arising from Table IV; the higher one gives 2.861, which is intermediate in terms of the experimental and adjusted values of Table XIV.

Because of these currently unresolved discrepancies, a second combination was calculated. For Adjustment B, the constant c was held at 2136.2, which corresponds to the experimental input from the product $\nu_t(^{239}\text{Pu})\sigma_f(^{239}\text{Pu})$. Applying greater weight to the experimental values of the absorption cross section and the neutron yield developed a smaller increase in capture. The decrease in η continues at 0.84% below the IAEA recommendations.

There are two reasons for giving slight preference to this latter arrangement. First, the various linear extrapolations of Colvin^{15,16} for ^{239}Pu $\nu_t(E)$ data are consistent with a 2200-m/s intercept of 2.854 neutrons/fission or less. Second, the experimental results of Magnuson⁵³ for the product $\eta(^{239}\text{Pu})\sigma_a(^{239}\text{Pu})/\eta(^{235}\text{U})\sigma_a(^{235}\text{U})$ are more in accord with a value $c = 2136.2$. The measured value of the $c(^{239}\text{Pu})/c(^{235}\text{U})$ ratio is 1.509 ± 0.025 , compared to 1.5075 now obtained from Adjustment B and 1.5197 from Adjustment A. The error attributed to the measurement has been increased by Hanna *et al.*⁴² because of spectral uncertainties and g-factors. A smaller error was reported by Magnuson.

IV. DISCUSSION

My independent scrutiny has resulted in a supportable hypothesis that significant systematic discrepancies exist among the accepted 2200-m/s fissile constants. The derived adjusted values typically differ by about three quoted standard deviations from the latest IAEA recommended list.⁴² Table XV contains estimates of the uncertainties in the major 2200-m/s fission parameters.

TABLE XV. Estimated Accuracies^a
of the Fission Parameters

	^{233}U	^{235}U	^{239}Pu		^{233}U	^{235}U	^{239}Pu
σ_a	$\pm 0.4\%$	$\pm 0.3\%$	$\pm 0.8\%$	η	$\pm 0.6\%$	$\pm 0.3\%$	$\pm 0.4\%$
σ_f	$\pm 0.4\%$	$\pm 0.4\%$	$\pm 0.4\%$	ν	$\pm 0.5\%$	$\pm 0.3\%$	$\pm 0.8\%$
σ_γ	$\pm 10\%$	$\pm 1.3\%$	$\pm 3\%$	σ_s	$\pm 20\%$	$\pm 11\%$	-
α	$\pm 7\%$	$\pm 1.3\%$	$\pm 3\%$				

^a67% confidence level.

A. Systematic Effects

The adjustments made in this report are clearly systematic. They arise from selection of a basis for experiment credibility that differs from that of the IAEA team. Credibility assessment is ultimately translated to weighting factors which determine the relative role of a datum within a set of diverse data. I have removed some of the downweighting applied to experiments which incorporate a variety of verification procedures through cross-checks and redundancy. Thus, particularly for the half-lives of ^{233}U and ^{234}U and for absolute measurements of ^{252}Cf neutron yield, an entirely different interpretation of the full complement of 2200-m/s values arises. The weighted average fission cross section for ^{235}U becomes higher, among

other effects. These experimental averages for $\sigma_f(^{235}\text{U})$ and the $\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$ ratio both agree nearly exactly with a completely independent evaluation by Deruytter.²⁴

A detailed examination of the ^{252}Cf experiments supports the validity of a lower ν_t . In addition, current manganese-bath practice indicates that two of the three precision measurements of η should have their values decreased.

In the reassembling of these data--retaining the general constraints of constancy of the products $\nu\sigma_f = \eta\sigma_a$ --the ^{235}U adjusted set does not differ much from the reassessed experimental averages, but they both differ markedly from the evaluations published in 1965 and 1969 by the IAEA. The most difficult feature of the adjusted set to support with external experimental data is the increased absorption cross section. For ^{235}U there is a difference of 1/2% between the reassessed experimental average and the adjusted value. It is shown in earlier sections that this difference is not implausible.

The larger differences in capture fractions are also not too difficult to accept in view of the history of previous α averages and in view of the nonmonochromatic nature of these measurements.

One should not consider the analytical procedure used in this report to be a substitute for the least-squares-fitting method; nor, conversely, can the statistical fit be considered valid without the extraction of what appear to be substantial systematic errors. If the inconsistencies evident in the prior IAEA input data were remedied, then--coupled with the remaining vast reservoir of unaffected input data--a revised least-squares fit should develop sufficient internal consistency to justify the derived errors. From a nonexperimental viewpoint, the next step recommended is an adjudication of the issues raised in this report, ultimately leading to a refitting of the data. However, it would be wise to proceed with the reassessment no faster than the availability of additional clarifying experimental data. Meantime, the values recommended in the present report can indicate the current probable values of the fundamental fission parameters at 2200 m/s and the degree of uncertainty associated with these parameters.

B. Role of Integral Measurements

I consider here some aspects of the past, present, and future role of integral reactor experiments. The conditions of reactor criticality have been responsible for the constraints that effectively keep the products $\nu\sigma_f = \eta\sigma_a$ a constant. Beyond this, though, there has been little extracted from the integral data because of the complications of heterogeneity, neutron escape, spectrum, g-factors, energy dependence of cross sections, and other features that confuse the interpretation of data in terms of 2200-m/s constants.

In fact, it is opportune to observe that integral measurements have failed to call attention to the significant discrepancies isolated in this report. The reactor critical experiment is usually not sufficiently sensitive at this level of precision for 2200-m/s constants.

A case in point is the measurement series by Gwin and Magnuson,⁴¹ treated by the authors and by the IAEA as an accurate determination of η : "Critical dimensions of both spherical and cylindrical volumes were measured as a function of the chemical concentration of the fissile isotope. In one of the spheres, the critical concentration was measured as a function of boron concentration." With the aid of some supplementary experiments and calculations, the critical condition

$$\bar{\eta}fFP(B) = 1 \quad (1)$$

was solved to derive $\bar{\eta}$, the spectrum-averaged neutron yield per absorption. In Eq. 1, f is the thermal utilization

$$f = \frac{\bar{\Sigma}_{ax}}{\bar{\Sigma}_{at}}, \quad (2)$$

where the numerator is the macroscopic absorption cross section of the fissile isotope, and the denominator is the total macroscopic absorption cross section of the entire solution; F is the fast-fission factor times the resonance escape probability, and

$$P(B) = \frac{K(B, E_{th})}{1 + L^2 B^2}, \quad (3)$$

where K is the nonleakage probability, L is the thermal diffusion length, and B^2 is the buckling.

Values of the thermal utilization ranged from 0.45 to 0.59, which means that about half the total absorptions occurred in the fissile isotope. Thus, aside from a limited possibility of fitting the system absorption ratio to the coefficient of a single parameter fit, the actual outcome of the experiment is dependent upon the input value of the absorption cross section of the fissile isotope--to the extent that $\Delta\eta/\eta \approx 0.5(\Delta\sigma_a/\sigma_a)$, as acknowledged by Gwin and Magnuson.⁴¹ We may consider this experiment just as much a measurement of $\eta\sigma_a$ as a measurement of η . To the limits of accuracy, these Oak Ridge critical experiments provide a constraint on the product $\nu\sigma_f = \eta\sigma_a = c$.

Three other factors emerge. First, it is evident why--within the limits of precision of such experiments--critical experiments are relatively insensitive to exchanges in the magnitude of η and σ_a . The Oak Ridge measurements are evaluated by the IAEA with total errors of about 1%, which is enough to disguise important changes.

Second, one must recall that considerable criticality data exist for ^{235}U thermal systems. To breach the constraining value of the constant c is likely to be inconsistent with this wealth of data.

Third, somewhat differently for ^{233}U and ^{239}Pu , the range of available thermal criticality data is more limited. Gwin and Magnuson carried out direct experiments on criticality of ^{233}U solutions, but the limited worldwide availability of the lower-weight fissile isotope does not permit as strong an inference to be drawn regarding corresponding knowledge of the constant $c(^{233}\text{U})$. Due to the relative paucity of moderated plutonium criticals, the status of the condition $c(^{239}\text{Pu})$ is even less established.

Consequently, in the absence of information to the contrary, a relatively high degree of confidence must be associated (as the IAEA, in its two surveys^{42,71}) with the boundary condition $c(^{235}\text{U})$. Descending confidence can be placed in $c(^{233}\text{U})$ and $c(^{239}\text{Pu})$. Or, to put it another way, violation of these conditions, for cause, is easier with ^{239}Pu than with ^{235}U . As a corollary, because reactor criticality experience is even less sensitive to the exact mix of either pair of parameters, $\eta\sigma_a$ or $\nu\sigma_f$, one can understand why the critical-mass data have not resolved the dilemma to date.

Future integral experiments probably cannot help resolve the general pattern of discrepancy. Perhaps assistance in choosing between the two adjusted sets of ^{239}Pu data may be obtained from examination of existing or newly generated reactor experiments.

It is reasonable to conclude that integral reactor measurements are unlikely to provide the major variations necessary to illuminate these 2200-m/s data discrepancies, particularly because of the high cost and restricted range of such integral measurements.

C. Differential Experiments Needed

To eliminate the uncertainty associated with the contradictory evaluations of the 2200-m/s constants, one must call upon assistance from direct differential experiments.

Further determinations of the ^{233}U and ^{234}U decay constants, especially by a variety of techniques and at different laboratories, would be of considerable value. The accurate measurements of CBNM¹⁹ are important in this regard.

Additional accurate measurements of the ν_t ratio for $^{239}\text{Pu}/^{252}\text{Cf}$ are essential in terms of clarifying the ambiguity in adjusted value for the important plutonium breeder-reactor fuel. The CEA team of Soleilhac et al.⁶⁴ is best equipped to find this ratio at or near 2200 m/s.

The forthcoming measurements for additional absolute values of $\nu_t(^{252}\text{Cf})$ by Axton⁴ are potentially important contributions. Also, supportive work regarding manganese-bath correction factors for ^{252}Cf sources is needed from a second laboratory.

The coexperimenters White and Axton⁷² can help clear up a muddled picture if they will make some efforts to directly reconcile their respective independent fission-fragment counting discrepancies.

The remaining problems regarding large liquid-scintillator measurements are best attacked in two ways: (1) by replication of initial conditions as much as possible at Los Alamos and at Stockholm in a systematic search for the delayed gamma and gamma-ray multiplicity effects; and (2) by another absolute liquid-scintillator measurement done with careful attention to these factors. Here again, the French group is best prepared to follow through for an absolute determination of ν_t for ^{252}Cf .

Another η measurement, especially if done by a different technique, would be useful in testing the 3/4% reduction called for in the data readjustment.

The NRU/NRX irradiation measurements to determine capture-to-fission and fission cross-section ratios need to be reexamined for possible inadequacy of correction factors. Little is likely to be gained from further irradiation measurements, as such, but much could be gained from supplementary studies.

There is only one specific measurement of the 2200-m/s scattering cross section for ^{235}U and none for ^{233}U and ^{239}Pu ; either additional work along these lines or some direct measurements of fissile absorption cross sections are desirable. A possible configuration for an absorption measurement would be by means of a high thermal-cross-section spherical-cavity detector, relatively insensitive to fission neutrons, surrounding fissile targets.

Finally, although not of direct bearing on the fissile-nuclide dilemma, an accurate value for the hydrogen thermal capture cross section would be useful.

V. CONCLUSIONS

I find strong evidence that the fundamental 2200-m/s fission parameters need significant revisions. The evidence is of two types. First, from interpretation of certain experiments at a level of credibility deviating from that in the IAEA review, a markedly different set of experimental averages arise for the neutron yield and for the fission cross sections. Second, there is substantive independent evidence that these new averages are valid. The

new combination of averages leads to an adjusted set of fission constants which differ typically by three standard deviations from the IAEA recommended values. The adjusted ^{235}U set was calculated by retaining the constraints that $\nu\sigma_f$ and $\eta\sigma_a$ equal the same constant value chosen by the IAEA. As a result, reactor integral experiments are not especially sensitive to this rearrangement of the fissile constants. However, calculations of breeding margin for future reactor systems are dependent on the fissile constant mix. For example, Greebler *et al.*³⁹ calculate that a 1% across-the-board uncertainty in $\nu_t(^{239}\text{Pu})$ produces a 1.5% effect on breeding ratio, corresponding to a much larger influence on doubling time. This 1% uncertainty also means a difference of 1.5% in fissile plutonium fuel inventory.

One of the more severe implications of this analysis is in terms of the prospects for thermal breeding in the ^{233}U - ^{232}Th cycle. There the extra neutron margin is so limited that the reduction in neutron yield, although compensated by increased fission probability and reduced parasitic capture, may be significantly deleterious. For a spectrum-averaged ^{233}U value of $\eta = 2.07$, the effect of a 0.86% decrease in 2200-m/sec η could be to increase the fuel doubling-time by nearly a factor of two. However, a second adjustment option, based on violation of the constraining $\eta\sigma_a$ product, is both more likely and less disruptive.

The revised 2200-m/s cross-section set developed in this report contains decreases in ν_t amounting to nearly 1% and slightly smaller reductions in η for all the fissile isotopes. To compensate for these changes, higher fission cross sections and reduced capture fractions are recommended.

Because of insufficient data, there is an additional uncertainty in the ^{239}Pu data.

The disintegration constants for ^{233}U and ^{234}U appear to be about 2% higher than the IAEA average.

To resolve the subjective aspects of the 2200-m/s cross-section fitting procedure, a series of sensitive differential experiments is advised.

REFERENCES

1. E. J. Axton, P. Cross, and J. C. Robertson, *Calibration of the NPL Standard Ra-Be Photoneutron Sources by an Improved Manganese Sulphate Bath Technique*, J. Nucl. Energy, Parts A/B 19, 409 (1965).
2. E. J. Axton and T. B. Ryves, *Scattering Resonance Self-Shielding Correction for Use with Manganese Sulphate Solutions*, J. Nucl. Energy 21, 543 (1967).
3. E. J. Axton, A. G. Bardell, and B. N. Audric, *A Technique for the Absolute Counting of Nuclear Fission Events*, J. Nucl. Energy 23, 457 (1969).
4. E. J. Axton, *Consultants' Meeting on $\bar{\nu}$* , IAEA Meeting, Studsvik, Sweden, June 10-11, 1970.
5. J. Barre', *Consultants' Meeting on α* , IAEA Meeting, Studsvik, Sweden, June 12, 1970.
6. C. B. Bigham, G. C. Hanna, P. R. Tunnicliffe, P. J. Campion, M. Lounsbury, and D. R. MacKenzie, "The Slow Neutron Fission Cross Sections of the Common Fissile Nuclides," in *Proc. 2nd UN Intern. Conf. Peaceful Uses At. Energy*, Geneva, 1958, Vol. 16, p. 125. United Nations, New York, 1965.
7. J. W. Boldeman and A. W. Dalton, *Prompt Nubar Measurements for Thermal Neutron Fission*, AEC/E-172 (Mar 1967).
8. M. M. Bretscher and W. C. Redman, *Low Flux Measurements of ^{239}Pu and ^{235}U Capture-to-Fission Ratios in a Fast Reactor Spectrum*, Nucl. Sci. Eng. 39, 368 (1970).
9. M. M. Bretscher, J. M. Gasidlo, and W. C. Redman, " Pu-239 , U-235 and U-238 Capture-to-Fission Ratios in ZPR-3 Assembly 57 Measured by the Reactivity-reaction Rate Method," in *Applied Physics Division Annual Report: July 1, 1969, to June 30, 1970*, ANL-7710, p. 119 (Jan 1971).
10. M. J. Cabell, "Harwell Mass Spectrometric Measurements of the Ratio of Neutron Capture to Fission for ^{233}U , ^{235}U , ^{239}Pu and ^{241}Pu in Reactor and Maxwellian Neutron Spectra," in *Proc. Conf. Nuclear Data for Reactors*, Paris, October 17-21, 1966. IAEA, Vienna, 1967.
11. G. Casini, private communication (June 23, 1970).
12. H. Ceulemans and F. Poortmans, "The Scattering Cross Section of ^{235}U between 0.025 eV and 1 eV," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
13. D. W. Colvin and M. G. Sowerby, "Boron Pile $\bar{\nu}$ Measurements," in *Proc. Symp. Physics and Chemistry of Fission*, Salzburg, March 22-26, 1965. IAEA, Vienna, 1965.
14. D. W. Colvin, M. G. Sowerby, and R. I. McDonald, "Confirmatory Experimental Data on the Harwell Boron Pile $\bar{\nu}$ Values," in *Proc. Conf. Nuclear Data for Reactors*, Paris, October 17-21, 1966. IAEA, Vienna, 1967.
15. D. W. Colvin, EANDC(68), p. 64 (July 3, 1968).
16. D. W. Colvin, "The Number of Neutrons per Fission, $\bar{\nu}$, from Thermal to 15 MeV," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.

17. H. Conde', $\bar{\nu}$ of ^{252}Cf , IAEA Panel on Neutron Standards, Brussels, May 1967.
18. H. Conde', *Consultants' Meeting on $\bar{\nu}$* , IAEA Meeting, Studsvik, Sweden, June 10-11, 1970.
19. P. De Bievre, K. F. Lauer, Y. Le Duigou, H. Moret, G. Muschenborn, J. Spaepen, A. Spennol, R. Vaninbrouckx, and V. Verdingh, *The Half-Life of ^{234}U* , EANDC(E)-133-AL (1970); also reported at EANDC Symp. on Neutron Standards and Flux Normalization, Argonne National Laboratory, October 21-23, 1970, to be published as CONF-701002.
20. J. A. De Juren and J. Chin, J. Res. Natl. Bur. Stand. 55, 311 (1955).
21. A. J. Deruytter, *The Fission Cross Section of ^{235}U from 0.01 eV to 0.1 eV and Its Absolute Value at 0.0253 eV*, J. Nucl. Energy, Parts A/B 15, 165 (1961).
22. A. J. Deruytter, J. Spaepen, and P. Pelfer, "Precise 2200 m/s Fission Cross Section of ^{235}U ," in *Proc. Conf. Neutron Cross Sections and Technology*, Washington, D.C., March 4-7, 1968. U.S. National Bureau of Standards Spec. Publ. 299, p. 491 (1968).
23. A. J. Deruytter and W. Becker, "Precise 2200 m/s Fission Cross Sections of ^{239}Pu and ^{235}U ," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
24. A. J. Deruytter, "Precision Low Energy Fission Cross-Sections," presented at EANDC Symp. on Neutron Standards and Flux Normalization, Argonne National Laboratory, October 21-23, 1970, to be published as CONF-701002.
25. A. De Volpi and K. G. Porges, "Direct and Absolute Measurements of Average Fission Neutron Yield from Uranium-235 and Californium-252," in *Proc. Conf. Nuclear Data for Reactors*, Paris, October 17-21, 1966, Vol. I, p. 297. IAEA, Vienna, 1967.
26. A. De Volpi, *Current Values of Fundamental Fission Parameters*, Reactor Fuel-process. Tech. 10(4), 271 (1967).
27. A. De Volpi, R. J. Armani, and K. G. Porges, *Extension of the Range of Determination of Manganese Sulphate Content for Neutron Source Measurements*, J. Nucl. Energy 21, 521 (1967).
28. A. De Volpi and K. G. Porges, *Absolute Calibration of Neutron Sources Having a Wide Range of Emission Spectra*, Metrologia 5, 128 (1969).
29. A. De Volpi and K. G. Porges, *Neutron Yield of ^{252}Cf Based on Absolute Measurements of the Neutron Rate and Fission Rate*, Phys. Rev. C 1, 683 (1970).
30. A. De Volpi (comp.), J. G. V. Taylor, A. Spennol, A. Rytz, B. J. Mijneer, A. Williams, and I. W. Goodier, *International Comparison of ^{56}Mn Activity in 1968*, Metrologia 6, 65 (1970); and ANL-7642 (Dec 1969).
31. A. De Volpi, *Neutron Escape from Water-Moderated Tanks*, J. Nucl. Energy 24, 577 (1971).
32. A. De Volpi, *Thermal Neutron Absorption in Neutron Sources and Cavity Walls Centered in Moderating Solutions*, to be published in J. Nucl. Energy (1971).

33. R. W. Durham, G. C. Hanna, M. Lounsbury, C. B. Bigham, R. G. Hart, and R. W. Jones, "Ratio of Capture to Fission in ^{235}U and ^{239}Pu ," in *Proc. Conf. Nuclear Data for Reactors*, Paris, October 17-21, 1966, p. 17. IAEA, Vienna, 1967.
34. P. A. Egelstaff, *The Harwell Values of the 2200 m/sec Neutron Data for U^{233} , U^{235} and Pu^{239}* , AERE-NP/R-2104 (Mar 18, 1957).
35. G. Frayssé and A. Prosdociimi, "Fission Cross-Section Measurement of ^{235}U and ^{239}Pu for Low-Energy Neutrons," in *Proc. Symp. Physics and Chemistry of Fission*, Salzburg, March 22-26, 1965. IAEA, Vienna, 1965.
36. W. J. Friesen, B. R. Leonard, and E. J. Seppi, *Comparison of a ^{235}U Fission Cross Section and ^{197}Au Capture Cross Section*, HW-47012 (Nov 1956).
37. V. F. Gerasimov and V. S. Zenkevich, *Atom. Energ.* 13, 368 (1962).
38. D. T. Goldman, P. Aline, R. Sher, and J. R. Stehn, *Twenty-two Hundred Meter per Second Neutron Absorption Cross Sections*, Bull. Am. Phys. Soc. 13, 1421 (1968).
39. P. Greebler, B. A. Hutchins, and C. L. Cowan, "Implications of Nuclear Data Uncertainties to Reactor Design," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
40. K. Gubernator and H. Moret, *Evaluation of the $^{10}\text{B}(n, \alpha)$ Cross Section and Branching Ratio*, EUR-3950e (1968).
41. R. Gwin and D. W. Magnuson, *The Measurement of Eta and Other Nuclear Properties of U^{233} and U^{235} in Critical Aqueous Solutions*, Nucl. Sci. Eng. 12, 364 (1962).
42. G. C. Hanna, C. H. Westcott, H. D. Lemmel, B. R. Leonard, Jr., J. S. Story, and P. M. Attree, *Revision of Values for the 2200 m/s Neutron Constants for Four Fissile Nuclides*, At. Energy Rev. 7(4), 3 (1969).
43. G. C. Hanna, private communication, Studsvik, Sweden (June 10-11, 1970).
44. J. C. Hopkins and B. C. Diven, *Prompt Neutrons from Fission*, Nucl. Phys. 48, 433 (1963).
45. H. R. Ihle, A. P. Murrenhoff, and M. Karayannis, "Standardization of Alpha Emitters by Liquid Scintillation Counting," in *Proc. Symp. Standardization of Radionuclides*, Vienna, October 10-14, 1966, pp. 69-76. IAEA, Vienna, 1967.
46. R. L. G. Keith, *The Half-Life of ^{233}U* , J. Nucl. Energy 22, 471 (1968).
47. R. L. G. Keith, A. McNair, and A. L. Rodgers, *A Measurement of the Fission Cross Sections of ^{233}U , ^{235}U and ^{239}Pu* , J. Nucl. Energy 22, 477 (1968).
48. C. J. Kenward, R. Richmond, and J. E. Sanders, *A Measurement of the Neutron Yield in Thermal Fission ^{235}U* , AERE R/R-212 (revised) (1958).
49. C. A. Kienberger, *The Uranium 234 Content of Natural Uranium and the Specific Alpha-Activities of the Isotopes*, Phys. Rev. 76, 1561-1563 (1949).
50. C. A. Kienberger, *The Specific Alpha-Activity of U^{234}* , Phys. Rev. 87, 520 (1952).

51. M. Lounsbury, R. W. Durham, and G. C. Hanna, "Measurements of Alpha and of Fission Cross Sections for ^{233}U , ^{235}U and ^{239}Pu at Thermal Energies," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
52. R. L. Macklin, G. De Saussure, J. D. Kington, and W. S. Lyon, *Manganese Bath Measurements of η of ^{233}U and ^{235}U* , Nucl. Sci. Eng. 8, 210 (1960).
53. D. W. Magnuson, *Values of Eta of Uranium-233, Uranium-235, and Plutonium-239 from Critical Experiments and Reactivity Measurements*, Nucl. Sci. Eng. 44, 266 (1971).
54. E. E. Maslin, J. A. Moore, J. M. A. Reichelt, and J. B. Crowden, *Absolute Fission Cross Section of ^{235}U for 2200 m/s Neutrons*, Phys. Rev. 139, B852 (1965).
55. D. Mather, *Consultants' Meeting on $\bar{\nu}$* , IAEA Meeting, Studsvik, Sweden, June 10-11, 1970.
56. J. W. Meadows, private communication (1970).
57. J. W. Meadows, "Thermal Capture Cross Sections of Li-6 and B-10 by the Pulsed Neutron Method," presented at EANDC Symp. on Neutron Standards and Flux Normalization, Argonne National Laboratory, October 21-23, 1970, to be published as CONF-701002.
58. J. A. Mitchell and C. J. Emert, "Evaluation of Eta for ^{233}U at 0.025 eV Using Monte Carlo," presented at 3rd Conf. on Neutron Cross Sections and Technology, Knoxville, Tennessee, March 15-17, 1971.
59. A. Moat et al., *Some Experimental Determinations of the Number of Prompt Neutrons from Fission*, J. Nucl. Energy, Parts A/B 15, 102 (1961).
60. F. L. Oetting (Ref. 94 quoted by Hanna et al.⁴²).
61. G. J. Safford, W. W. Havens, Jr., and B. M. Rustad, *A Precise Determination of the Total Cross Section of Uranium 235 from 0.000818 eV to 0.0818 eV*, Nucl. Sci. Eng. 6, 433 (1959).
62. A. Saplakoglu, "Absolute Thermal Fission Cross Section Measurements of ^{235}U by a New Method," in *Proc. 2nd UN Intern. Conf. Peaceful Uses At. Energy*, Geneva, 1958, Vol. 15, p. 103. United Nations, New York, 1958.
63. J. R. Smith, S. D. Reeder, and R. G. Fluhrty, *Measurement of the Absolute Value of Eta for U-233, U-235 and Pu-239 Using Monochromatic Neutrons*, IDO-17083 (1966).
64. M. Soleilhac, J. Frehaut, J. Gauriau, and M. Mosinski, "Measurement of Mean Number of Prompt Neutrons and Relative Cross Sections for the Fission of U-235 and Pu-239 by Neutrons with Energies between 0.3 and 1.4 MeV," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
65. A. Staub, D. R. Harris, and M. Goldsmith, *Analysis of a Set of Critical Homogeneous U-H₂O Spheres*, Nucl. Sci. Eng. 34, 263 (1968).
66. N. M. Steen, "Thermal Parameters for ^{233}U ," presented at 3rd Conf. on Neutron Cross Sections and Technology, Knoxville, Tennessee, March 15-17, 1971.

67. C. E. Till, J. M. Gasidlo, E. F. Groh, L. G. LeSage, W. R. Robinson, and G. S. Stanford, *Null-Reactivity Measurements of Capture/Fission Ratio in ^{235}U and ^{239}Pu* , Nucl. Sci. Eng. 40, 128 (1970).
68. C. A. Uttley and K. M. Diment, "Analysis of the Total Cross Section of Carbon from 70 eV to 1.5 MeV," presented at EANDC Symp. on Neutron Standards and Flux Normalization, Argonne National Laboratory, October 21-23, 1970, to be published as CONF-701002.
69. R. Vidal, M. Robin, and C. Carneiro da Silva, "Mesure des Sections Efficaces de L'Uranium 233 Pour les Neutrons Thermiques," in *Proc. 2nd Intern. Conf. Nuclear Data for Reactors*, Helsinki, June 15-19, 1970. IAEA, Vienna, 1971.
70. R. B. Walton and R. E. Sund, *The Effect of Fission-Delayed Gamma Rays on X Measurements of ν_p Performed with Large Liquid Scintillation Detectors*, Nucl. Instr. Methods 68, 163 (1969).
71. C. H. Westcott, K. Ekberg, G. C. Hanna, N. J. Pattenden, S. Sanatani, and P. M. Attree, *A Survey of Values of the 2200 m/s Constants for Four Fissile Nuclides*, At. Energy Rev. 3(2), 3 (1965).
72. P. H. White and E. J. Axton, *Measurement of the Number of Neutrons per Fission for ^{252}Cf* , J. Nucl. Energy 22, 73 (1968).
73. P. H. White, *Alpha and Fission Counting of Thin Foils of Fissile Material*, Nucl. Inst. Methods 79, 1 (1970).

ARGONNE NATIONAL LAB WEST



3 4444 00010924 9